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### Generation of normal and adakite-like calc-alkaline magmas in a non-subductional environment: An Sr-O-H isotopic study of the Apuseni Mountains neogene magmatic province, Romania

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**GENERATION OF NORMAL AND ADAKITE-LIKE CALC-ALKALINE  
MAGMAS IN A NON-SUBDUCTIONAL ENVIRONMENT: A Sr-O-H ISOTOPIC  
STUDY OF THE APUSENI MOUNTAINS NEOGENE MAGMATIC PROVINCE,  
ROMANIA**

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***Abstract***

Neogene magmatism in the Apuseni Mountains of Romania was active between 15 and 7 Ma, with an isolated final eruption at 1.6 Ma. Several groups of magmatic rocks can be distinguished in the province on the basis of ages, trace elements and isotopic compositions. Most are of calc-alkaline affinity, despite the fact that the region is located 200 km from the postulated Carpathian subduction trench. However, adakite-like calc-alkaline magmas were also erupted and the youngest eruptions were of alkaline affinity. Magmas older than 13.5 Ma are characterised by the highest  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values in their phenocryst phases. This early magmatism is considered to be a result of crustal melting related to the fast rotation of small crustal blocks. Volcanic rocks younger than 13.5 Ma have lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  ratios, with a narrow range for pyroxene and

amphibole mineral separates, indicating closed system fractional crystallization. A larger scatter of  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  values is found in plagioclase phenocrysts and groundmasses, indicating that a small amount of crustal assimilation also occurred. The  $\delta\text{D}$  values for hydrogen-bearing phases such as amphibole, biotite and groundmass indicate fractional crystallisation in a closed system and a trend of decreasing  $\delta\text{D}$  in the source with time. The magma source of normal calc-alkaline adakite-like magmas was not related to contemporaneous subduction, but was likely due to melting of delaminated eclogitic lower continental crust. In contrast, the late-stage alkaline magmas were related to asthenospheric upwelling. Petrological analysis of the complex succession of magmas in the Apuseni Mountains province demonstrates that normal calc-alkaline and adakite-like magmatism can be generated in a non-subductional environment, in this case associated with an unusual extensional setting.

**Key words:** Carpathian-Pannonian region, Apuseni Mountains, magmagenesis,  $^{87}\text{Sr}/^{86}\text{Sr}$ , stable isotope geochemistry.

## ***Introduction***

The Apuseni Mountains magmatic province in western Romania forms part of the wider area of Neogene calc-alkaline volcanism of the Carpathian-Pannonian region (Fig. 1). The province is situated at a distance of 200 km from the inferred subduction trench, which suggests that processes other than subduction must have been involved in magma generation. Apuseni magmatism consists of pyroxene-, amphibole- or biotite-bearing andesites with a few basaltic andesites, dacites and rare garnet-bearing andesites, developed in four NW-SE trending volcano-intrusive areas (Roşu *et al.*, 2004a). The largest continuous outcropping area is ca. 100 km long (Fig. 1). Magmatic activity occurred from 14.7 to 7.4 Ma and concluded with a brief period of trachyandesitic volcanism at ~1.6 Ma (Pécskay *et al.*, 1995; Roşu *et al.*, 1997, 2004a).

Roşu *et al.* (2001, 2004a) and Seghedi *et al.* (2004) recently recognized that some magmatic rocks in the Apuseni region have unusual geochemical characteristics that are

similar to those of adakites. Samples from the province can thus be classified into normal calc-alkaline, adakite-like calc-alkaline and alkaline types (Roşu *et al.*, 2004a). Magmatism in the Apuseni Mountains has been interpreted by Seghedi *et al.* (1998, 2004) and Roşu *et al.* (2004a) as being due to extension that caused magma generation by melting of the upper lithospheric mantle or lower continental crust, later followed by asthenospheric mantle upwelling. However, the extent of crustal contamination and fluid involvement in the genesis of the Apuseni magmas is still unknown. For this purpose, we have analysed O, H and Sr isotopes in fresh phenocrysts and groundmass from volcanic rocks that represent the entire spectrum of lava compositions and the whole time interval of magma generation in the Apuseni province, for which whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  isotope data were already available (Table 1).

Oxygen and hydrogen isotope ratios, in combination with  $^{87}\text{Sr}/^{86}\text{Sr}$ , provide powerful constraints on magmagenesis, particularly the extent of crustal contamination, as well as the possible effects of fluid involvement. In this paper we report 27 new oxygen isotope data, 15 hydrogen and 18 new Sr isotope analyses on mineral separates and groundmass from eight samples from the Apuseni Mountains. We have analysed oxygen isotopes on separated minerals because several andesitic rock series from elsewhere in the world (e.g. Downes *et al.*, 1995; Downes *et al.*, 2001) have shown a significant difference between the  $\delta^{18}\text{O}$  values obtained for mafic minerals and those of the whole-rock powder, with the differences ascribed to low-temperature alteration. A unique feature of this study is the application of Sr isotope analyses to separated phenocrysts and groundmass in the volcanic rocks, in an effort to avoid the effects of late-stage alteration that may be associated with whole-rock Sr analyses. It has been shown by Davidson *et al.* (1998) and Tepley *et al.* (2000) that Sr isotope analyses of plagioclase and groundmass can provide insights into processes that affected the magmas in the time interval during which the phenocrysts and groundmass were formed.

### ***Tectonic setting of the Apuseni Mountains***

The basement of the Carpathian-Pannonian region comprises an assemblage of different continental blocks, with the Alcapa block in the north and the composite Tisia-

Dacia block in the south (Balla, 1984, Csontos *et al.*, 1992; Brezsnysky *et al.*, 2000; Neugebauer *et al.*, 2001). Tisia and Dacia are microplates with different Mesozoic tectonic histories (Surmont *et al.*, 1990; Pătrașcu *et al.*, 1994) that collided in Early to Mid-Cretaceous times (Balintoni 1997; Dallmeyer *et al.*, 1999; Iancu *et al.*, 2005). The Apuseni Mountains are outcrops of the Tisia terrane (Kazmer, 1986; Bleahu *et al.*, 1996). Tectonic reconstructions suggest that, during Early to Middle Miocene times, both the Tisia-Dacia and Alcapa blocks experienced eastward translation (Csontos, 1995; Fodor *et al.*, 1999; Huisman *et al.*, 2001), synchronous with subduction retreat, collision and orogenesis in the East Carpathians (Royden, 1988; Săndulescu, 1988; Royden and Burchfiel, 1989; Csontos *et al.*, 1992; Csontos, 1995).

During Eocene to Early Miocene times, the Tisia-Dacia block experienced  $\sim 20^\circ$  clockwise rotation (Pătrașcu *et al.*, 1994; Panaiotu, 1998) at the same time as the Alcapa block underwent counterclockwise rotation (Márton and Márton, 1996). The northeastern part of Alcapa was subject to continuous rigid counterclockwise rotations of  $20^\circ$ - $50^\circ$  (Panaiotu, 1998; Márton *et al.*, 2000) at around 14 Ma to 12 Ma. At the same time, the Apuseni Mountains underwent very fast clockwise rotation ( $\sim 60^\circ$ ) (Panaiotu, 1998, 1999), whereas a neighbouring area on the Tisia-Dacia block, the Mecsek-Villany area, did not rotate. The large clockwise rotation of the Apuseni part and non-rotation of the Mecsek-Villany area took place across strike-slip faults (Csontos *et al.*, 2002). As a result, extension with a NW-SE strike occurred along the western border of the Apuseni Mts. (Royden, 1988; Săndulescu, 1988; Balintoni and Vlad, 1998). In addition, extension-related basins such as the Bekes and Zarand basins were generated in adjacent areas beneath the Great Hungarian plain (Balla, 1984; Györfi and Csontos, 1994). Sedimentation and magmatism in the Apuseni Mountains began at 16-15 Ma (Ghițulescu and Socolescu, 1941; Balintoni and Vlad, 1998; Roșu *et al.*, 1997, 2001, 2004a; Seghedi *et al.*, 1998, 2004). A consequence of this rotation was the synchronous extensional (Györfi and Csontos, 1994; Tari *et al.*, 1999) and compressional deformation (Huisman *et al.*, 1997; Ciulavu *et al.*, 2000), respectively west and east of the Apuseni Mts. During the Late Miocene to Recent, stress indicators for the southern part of Tisia and its rims show that an E-W to NW-SE compressive stress prevails, along with important isostatic

uplift (Ratchbacher et al., 1993; Bojar et al., 1998; Ciulavu *et al.*, 2000; Huismans *et al.*, 2001; Sanders *et al.*, 2002).

Figure 1 also shows an integrated interpretation of low-frequency seismic reflection along the PGT4 profile with regional geothermal, geomagnetic, gravity and magnetotelluric data at the western margin of the Apuseni Mountains (Takács *et al.*, 1996) that shows an asthenospheric upwelling below the Zarand basin, the largest graben system at the western edge of the Apuseni Mountains.

Figure 2 shows interpretations by Csontos *et al.* (2002) and Seghedi *et al.* (2004) of the geodynamic situation after the end of the differential rotation of the central and eastern parts of Tisia-Dacia, synchronous with graben opening in the western part of the Apuseni Mountains. The studied magmatism is closely connected with the major eastward rotations between 15.5 and 11 Ma and subsequent opening of narrow graben-type basins. During this interval, contemporaneous calc-alkaline magmatism also developed along the Carpathian arc in the front of Alcapa and Tisia-Dacia (Pécskay *et al.*, 1995, 2006), in direct relationship with rollback subduction retreat and probable breakoff of the subducted slab (Nemčok *et al.*, 1998; Seghedi *et al.*, 1998, 2001, 2004; Wortel and Spackman, 2001). During Late Middle Miocene times (~11 Ma), collision of Tisia-Dacia with the East European platform took place, causing the retreating subduction processes to cease (Csontos, 1995; Mañenco, 1997; Zweigel, 1997).

### ***Magmatic development of the Apuseni Mountains Province***

A complete discussion of the geochemistry of the magmatic rocks under consideration is given by Roşu et al. (2004a). The studied samples all contain 55-63 wt% SiO<sub>2</sub> and only two (776 and UR-3) have conspicuously high K<sub>2</sub>O contents. Several temporal trends can be seen in the major element, trace element and isotopic compositions. Total alkalis, P<sub>2</sub>O<sub>5</sub>, MgO and <sup>144</sup>Nd/<sup>143</sup>Nd ratios increase with time, whereas <sup>87</sup>Sr/<sup>86</sup>Sr ratios decrease. Additionally, Nb/Y and Sr/Y ratios show a general increase from the onset to the end of magmatic activity (Fig. 3). These trends suggest a tendency towards more alkaline magmatism and more “primitive” magma sources. Each period of eruption was characterized by specific magma compositions. Rocks older than

12.5 Ma are normal calc-alkaline; those younger than 12.5 Ma show high Sr/Y ratios, suggesting an adakitic nature; those generated between 10 and 7.4 Ma have higher Nb contents and Nb/Y ratios; the young alkaline volcano (1.6 Ma) shows a trend towards an intraplate OIB-type composition (Fig. 3).

Since among the normal calc-alkaline rocks, the older ones (14.5-13.0 Ma) have higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, Roşu *et al.* (2004a) suggested that, in the early stages of magma generation, either a larger amount of assimilation-fractional crystallization (AFC) occurred or that the source had a variable  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio. In contrast, fractionation processes were less significant for the younger magmatic products: they show a narrow  $\text{SiO}_2$  interval, no Eu anomalies, and have lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and higher  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios than the older ones. For rocks younger than 12 Ma, fluid involvement was more significant during magma generation, indicating the onset of the conditions that led to the generation of adakite-like magmas. Variable Ba/La ratios, higher for magmas in the SE areas, suggest a heterogeneous source. The observed temporal changes in magma composition may have resulted from changes in the composition of the local magma source, possibly of lower crust and/or mantle lithosphere origin and, in the late stages, involvement of the asthenosphere (Roşu *et al.*, 2004a).

### ***Sample selection and analytical techniques***

Rock types, geographic locations and key geochemical characteristics of samples examined in this study are given in Table 1. Petrographic and compositional information can be found in Roşu *et al.* (2004a). The rocks were first crushed to 0.30 mm in a percussion mortar. Rock chips were sieved and handpicked under a binocular microscope for fresh phenocrysts free of glass or alteration. We concentrated clinopyroxene, amphibole, biotite and plagioclase phenocrysts and also separated groundmass material composed of glass and various microliths. Clean phenocrysts were ultrasonically washed in acetone and dried prior to analysis.

Electron microprobe analyses of pyroxenes were carried out using a Jeol 8600 Superprobe at the Department of Earth Sciences, Utrecht University. Amphiboles from samples 776 and 767 were also measured. All measurements were performed using WDS

with an accelerating voltage of 15 kV, a beam current of 10 nA and spot sizes of 10  $\mu\text{m}$ . Counting times were 30 seconds for Mg, Al, Si, and Ca, 50 s for Ti and Fe, 100 s for K and 200 s for P and Mn, and 40 s for Na. For each rock, between 10 and 20 pyroxene crystals (as mineral separates mounted in epoxy resin) have been analysed and Table 2 gives a selection of the most representative crystal compositions.

Oxygen isotope analyses of minerals and groundmass were carried out at the Institute of Earth Sciences, Geology and Paleontology, University of Graz. Samples were heated with a 20W  $\text{CO}_2$  laser following the technique of Sharp (1990). Oxygen was extracted from silicate minerals by fluorination with  $\text{BrF}_5$  and was measured directly on a Finnigan MAT Delta Plus mass spectrometer without combustion to  $\text{CO}_2$ . Throughout the measurements several standards were analyzed together with the samples. For standards and samples 1–2 mg of material were analyzed. The reproducibility of measurements was tested on UWG-2 garnet standard (Valley et al., 1995; mean value of 5.74 ‰ and a standard deviation of 0.15 ‰) for which a mean value of 5.8 ‰ and a standard deviation of 0.15 ‰ were obtained. Measurements on NBS 30 biotite gave an average value of 5.04 ‰ and a standard deviation of 0.2 ‰ (accepted value 5.1 ‰, standard deviation 0.2 ‰). Hydrogen isotope measurements were done at the Department of Earth and Planetary Sciences, Albuquerque, following the technique of Sharp *et al.* (2001). The water content was calculated from the intensity of the mass 2 signal. All the  $\delta^{18}\text{O}$  and  $\delta\text{D}$  data are expressed relative to SMOW and are shown in Table 3. Sr isotope analyses were also performed on a subset of minerals and groundmass from the same samples. 3–15 mg of sample was dissolved in Savillex screw-top beakers using a mixture of HF and  $\text{HNO}_3$ . Sr was separated using Sr-spec Eichrom resin. Two total procedural blanks at the beginning and end of sample analysis gave values of about 0.14ng of Sr. Measurements were made using a Finnigan MAT 262 multicollector thermal ionization mass spectrometer at the University of Pisa, Italy. Measured  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios were normalised to  $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ . Errors on a single measurement are the standard deviation of the mean ( $2\sigma_m$ ) and are in the region of 0.00001. Replicate analyses of the international reference standard NIST 987 gave an average value of  $^{87}\text{Sr}/^{86}\text{Sr} = 0.710242 \pm 0.000013$  (2SD). Results are given in Table 3.

## ***Results***

All the analyzed rocks lack olivine but contain phenocrysts of clinopyroxene, amphibole, biotite and abundant plagioclase. With the exception of andesite sample 5199, where the clinopyroxene Mg# is 78-80, all the clinopyroxenes show high Mg#s (87-91) (Table 2). Some rocks (e.g. 767, 788 and UR-3) contain a few crystals that show compositional differences between their rounded cores and their euhedral margins. This is most likely due to magma mixing since the crystal margins show a much higher Mg# than the cores, a feature that is incompatible with fractionation. In sample 788 one crystal has a rim with a lower Mg# than the core, which can be related to fractionation; however the rest of the crystals in this sample show high Mg#s. Amphiboles from samples 776 and 767 show Si contents (TSi > 6.5 apfu) characteristic of magnesiohornblende.

In Fig. 4a, the  $\delta^{18}\text{O}$  values of the mineral separates (Table 3) are plotted against  $\text{SiO}_2$  content of the whole-rocks (Table 1). Clinopyroxene phenocrysts in all the measured rocks show an extremely restricted range of  $\delta^{18}\text{O}$  values between 5.5 and 5.6 ‰. These values are slightly higher than the  $\delta^{18}\text{O}$  values for clinopyroxene phenocrysts in alkali basalts from the Carpathian-Pannonian region, which show a range between 5.1 and 5.3 ‰ (Dobosi *et al.*, 1998), but they are similar to  $\delta^{18}\text{O}$  values of clinopyroxene in mantle peridotite xenoliths worldwide (5.7 ‰; Matthey *et al.*, 1994). There is no correlation between whole-rock  $\text{SiO}_2$  content and pyroxene  $\delta^{18}\text{O}$  values and also no variation of the clinopyroxene  $\delta^{18}\text{O}$  values with age of the host rock. Some of the amphibole and biotite separates show similar or slightly higher  $\delta^{18}\text{O}$  values than the clinopyroxene (5.6-5.9 ‰ in samples 363, 767), but others are 0.8-1.8 ‰ higher (6.4-7.3 ‰ in samples 790, 776 and 401). Mineral separates from all samples show a shift toward higher  $\delta^{18}\text{O}$  values for plagioclase and groundmass. Plagioclase phenocrysts show a wide variation of  $\delta^{18}\text{O}$  values between 6.3 and 7.6 ‰. Even higher  $\delta^{18}\text{O}$  values are given by the separated groundmass (6.7-10.1 ‰). The values for the groundmass are higher for the highly porphyritic older rocks (8.0-10.1 ‰) compared to the younger and only slightly porphyritic basaltic andesite (788) and trachyandesite (UR-3) that yield values of 6.7 and 7.2 ‰, respectively (Table 1). Figure 4b shows  $\delta^{18}\text{O}$  mineral compositions plotted

against  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of the whole-rock. Most of the samples with low  $\delta^{18}\text{O}$  values in their clinopyroxenes also have low whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, but two samples that do not contain clinopyroxene (790 and 401) have elevated  $^{87}\text{Sr}/^{86}\text{Sr}_{\text{WR}}$  ratios (0.708 and 0.7056, respectively). Sample 790, representing the oldest magmatism at 14.6 Ma, contains amphibole with an elevated  $\delta^{18}\text{O}$  value of 6.5 ‰. In the case of 401, however, amphibole yields a value of 5.6 ‰, similar to that of clinopyroxenes from the other samples.

A comparison of the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in whole-rocks (Table 1), mineral separates and groundmasses (Table 3) reveals that the whole-rocks tend to have slightly higher values than amphibole or clinopyroxene separates, but similar values to the groundmass (where analysed). Most mafic minerals show a narrow range of  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$ , but plagioclase and groundmass show a shift toward both higher  $^{87}\text{Sr}/^{86}\text{Sr}$  and higher  $\delta^{18}\text{O}$  values. The plot of mineral  $^{87}\text{Sr}/^{86}\text{Sr}$  vs. mineral  $\delta^{18}\text{O}$  values (Fig. 4c) shows consistently lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for pyroxenes and amphiboles than the whole-rock values, with minerals separated from samples 776, 788, 767 and UR-3 having a rather restricted range (around 0.7042) and 401, 5199 and 363 having higher values (0.7044-0.7046). However, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for mafic minerals from samples 363 and 5199 are close to the whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio. The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for plagioclases are, with the exception of sample 5199, higher than those of the groundmass. Similar results have been obtained for volcanic rocks from El Chichón volcano (Mexico) by Tepley et al (2000). Clearly, the whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio should be the weighted mean of the values for the separated minerals and groundmass, and Figure 4c demonstrates that mafic minerals yield a clearly more primitive value (i.e. closer to that of the magmatic source) than that of the whole rock. This effect is particularly pronounced in samples 401 and 790, in which the difference between the whole rock value and that of the amphibole phenocrysts is  $>0.001$ .

The  $\delta\text{D}$  values on groundmass material vary from  $-84$  ‰ to  $-127$  ‰ (Table 3). They show a good correlation with the  $\text{SiO}_2$  and age of the rocks, the older rocks having higher  $\delta\text{D}$  values (Figs. 5a and 5b). The amphiboles and biotites show a similar pattern. Generally, within a single sample, the groundmass has the lowest values and the biotite and amphiboles the highest. Water contents in groundmass material range from 0.8 to 3.6

wt %, although in the youngest sample UR-3 the water content was below detection limit. Amphiboles and biotites have water contents in the range of 2-4.4 wt%. Results for amphiboles are similar to those for amphibole from calc-alkaline rocks elsewhere in the Carpathian-Pannonian region reported by Demeny *et al.* (2006) who suggested that both high and low  $\delta D$  were produced by degassing ( $H_2O$  or  $H_2$  release).

### ***Discussion of Isotopic Variation***

Fractional crystallization in a closed system induces  $\delta^{18}O$  crystal-melt fractionations of  $<1$  ‰ for oxygen and  $<20$  ‰ for hydrogen (DePaolo, 1981). For unaltered volcanic rocks the  $\delta^{18}O$  values vary between 5.5 to 10 ‰, the isotopic values of coexisting phases showing the following order of isotopic enrichment due to fractional crystallisation: biotite-hornblende-feldspar-groundmass (Taylor and Sheppard, 1986).

Some Apuseni samples (e.g. 790, 5199, 788, UR-3) show differences in  $\delta^{18}O$  values between clinopyroxene, amphibole, biotite, plagioclase and groundmass of less than 2 ‰ (Table 3), indicating isotopic fractionation within the expected range for fractional crystallization (Taylor and Shepard, 1986). Samples 767, 776 and 363 show up to 2 ‰ differences in  $\delta^{18}O$  values of their constituent minerals but all these samples have  $\Delta^{18}O_{\text{pyroxene-groundmass}}$  or  $\Delta^{18}O_{\text{amphibole-groundmass}}$  greater than 2 ‰. This suggests that either the groundmass is more altered than the mafic minerals, or that enrichment in  $^{18}O$  occurred in the magma between crystallisation of the phenocrysts and quenching of the groundmass. Sample 401 shows a fractionation factor  $\Delta^{18}O_{\text{amphibole-plagioclase}}$  of 2 ‰; in this case a mechanism other than fractional crystallization must have operated.

Combining the  $^{87}Sr/^{86}Sr$  and  $\delta^{18}O$  values, significant trends are evident (Fig. 4c). Both  $^{87}Sr/^{86}Sr$  and  $\delta^{18}O$  ratios of the amphibole of the oldest sample (790) show more enriched values that are probably derived from a more “crustal” source. The slightly higher  $^{87}Sr/^{86}Sr$  of the groundmass of 790 could indicate a small amount of assimilation of material with higher  $^{87}Sr/^{86}Sr$  but with similar  $\delta^{18}O$  values. Therefore it seems that, in addition to fractional crystallization, a small amount of crustal contamination also occurred. The  $^{87}Sr/^{86}Sr$  ratio for sample 401 is intermediate between that of 790 and all

the other samples; this may also imply some degree of crustal contamination. This sample is characterized by a large difference in  $^{87}\text{Sr}/^{86}\text{Sr}$  between amphibole and plagioclase and also a quite large  $\Delta^{18}\text{O}_{\text{amphibole-plagioclase}}$  of 2 ‰. This could indicate assimilation of material with higher  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  during and after crystallisation of plagioclase. All the other samples discussed in this paragraph fall in the group with generally low  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios; differences between them mainly concern the evolution of  $\delta^{18}\text{O}$  values of the different phases. For sample 767, there is a large difference in  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios between pyroxene and plagioclase, although the groundmass falls in the group with low values for  $^{87}\text{Sr}/^{86}\text{Sr}$ . This implies a similar assimilation mechanism as for sample 401, although the amount of assimilated material was lower. Samples 401 and 767 have distinctly higher modal plagioclase contents, so their higher  $^{87}\text{Sr}/^{86}\text{Sr}_{\text{plag}}$  as compared to the groundmass points towards assimilation during plagioclase crystallization. Tepley et al. (2000) suggested that the higher  $^{87}\text{Sr}/^{86}\text{Sr}_{\text{plag}}$  compared with groundmass in Mexican volcanic rocks were due to recharge by a higher-temperature magma with a lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio and higher Sr concentration.

Samples 776 and 363 show similar  $^{87}\text{Sr}/^{86}\text{Sr}$  values between phases, although when considering the groundmass, they show a difference of  $> 2$  ‰ in  $\delta^{18}\text{O}$ . Samples 5199, 788 and UR-3 show similar values for  $^{87}\text{Sr}/^{86}\text{Sr}$  for all the phases and  $\delta^{18}\text{O}$  variations  $< 2$  ‰ between different phases. This suggests that fractional crystallization in a closed system was the major process that formed these rocks.

For several Apuseni Mountains samples (776, 767, 401 and 363), the differences between the  $\delta\text{D}$  values of amphibole, biotite and groundmass are very close to 20 ‰ (Table 3), indicating fractionation during crystallization in a closed system for this element. The magmas thus evolved in a closed system regarding hydrogen, although Sr and O isotopes for some of the samples indicate possible assimilation. In general the samples show a temporal trend, with higher  $\delta\text{D}$  values characterizing the older samples and lower  $\delta\text{D}$  values for the younger ones (Fig. 5b). Accordingly, we can interpret the variation of  $\delta\text{D}$  values between samples as due to variation in  $\delta\text{D}$  of the source, with a more depleted source for the younger rocks. As well,  $\delta\text{D}$  data against the whole rock

SiO<sub>2</sub> contents show a very good positive correlation that may be interpreted as due to magma evolution (AFC) or mixing of melts with different source compositions (Fig. 5a).

Two phases of fluid participation can be discerned in the studied samples: (1) an early hydrous magmatic phase that gave rise to hydroxyl-bearing minerals such as amphibole and biotite, which show low  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios similar to those of clinopyroxene; (2) a later melt phase with increasingly higher  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$ , as suggested by the late crystallising phases (especially plagioclase and groundmass), indicating an effect of crustal assimilation. The restricted range of  $\delta\text{D}$  values within an individual rock, as well as the general low water contents of the groundmass do not support the involvement of an external fluid. The data are in agreement with the significant amount of assimilation-fractional crystallization (AFC) suggested for the early stages of magma generation (Roşu *et al.*, 2001, 2004a) and may suggest that, for some locations, low  $\delta^{18}\text{O}$  minerals have been removed and the residual liquid has also had its  $\delta^{18}\text{O}$  increased via assimilation. Low pressure-high temperature reactions with crustal xenoliths at different depths, reported from basaltic andesites from the Apuseni Mountains by Har (2005), may be responsible for the increase of  $\delta^{18}\text{O}$  of the groundmass during magma transit through the crust. In the Apuseni Mountains various late-stage hydrothermal phases that operated temporally or regionally have been responsible of widespread metallogenetic activity (Udubaşa *et al.*, 2003, Roşu *et al.*, 2004a, 2004b, Neubauer *et al.*, 2005), but the rocks in this study were unaffected by hydrothermal fluids.

### ***Petrogenetic processes – Comparison with neighboring areas***

In this section, the Apuseni Mountains data will be compared with  $\delta^{18}\text{O}$  data for phenocrysts in magmatic rocks from other volcanic areas in the Carpathian-Pannonian region in order to evaluate the role of various petrogenetic processes and to distinguish between possible magmatic sources. Figure 6 shows the variations of  $\delta^{18}\text{O}$  of pyroxene, amphibole and biotite versus whole-rock SiO<sub>2</sub> content for volcanic rocks from several different areas of the Carpathian-Pannonian region, such as the Călimani and South

Harghita volcanic areas of eastern Romania and the Central Segment volcanic area (as defined by Seghedi *et al.*, 2004) that includes volcanic fields in eastern Hungary, eastern Slovakia, southeast Ukraine and northern Romania. For most of the Apuseni Mountains samples, the low degree of  $^{18}\text{O}$  isotopic variation over the range of  $\text{SiO}_2$  can be explained by closed system fractional crystallization, although magma mixing has also been involved, as shown by the variation in clinopyroxene chemistry (Table 2). In the data for Călimani and the Central Segment volcanic areas, magmas show a very different trend of increasing  $\delta^{18}\text{O}$  values with increasing whole-rock  $\text{SiO}_2$  contents, which suggests that AFC took place during the early phases, probably during residence in small shallow magma chambers (Mason *et al.*, 1996, Seghedi *et al.*, 2004). In contrast, magmas from the South Harghita volcanic area show similarities with the Apuseni magmas, with little variation in  $\delta^{18}\text{O}$  values over a wider range of  $\text{SiO}_2$  contents, however at higher overall  $\delta^{18}\text{O}$  values.

The plot of the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for the mafic minerals against  $\delta^{18}\text{O}$  mineral values (Fig. 7) is significant for detection of the various contamination contributions in magmagenesis. Three possible contamination mechanisms have been invoked in the Carpathian-Pannonian region (Harangi *et al.*, 2006): (1) source contamination by addition of subduction components derived from a descending slab and its associated sediments, based on the increase of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (Pearce, 1982), (2) crustal contamination of mantle-derived magmas acquired via assimilation-fractional crystallization within crustal magma chambers based on the shift to higher  $\delta^{18}\text{O}$  values (James, 1981; Davidson *et al.*, 1990), and (3) mixing between mantle-derived magmas and melts derived from metasedimentary crust (Harangi *et al.*, 2001). Our modelling focuses on combined crustal assimilation-fractional crystallization (AFC), source contamination (SC) and mixing with metasedimentary lower crust (MLC), which must be quantified and separated before an attempt can be made to constrain the magmatic sources (e.g. mantle, slab-derived melts, crust) and to distinguish between different mantle (asthenospheric, lithospheric) and crustal sources. Results for all three models are shown in Figure 7.

Low  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for volcanic rocks in the Apuseni Mountains generally indicate a low degree of source enrichment, which does not support the operation of subduction processes. Moreover, the data (with the exception of samples 776

and UR3, which have the highest K<sub>2</sub>O contents) fall in the same field as primitive oceanic arc lavas (e.g. Eiler *et al.*, 2000), revealing that the range of  $\delta^{18}\text{O}$  is not evidently associated with “slab-derived fluid” (high Ba/La) or “sediment-derived melt” (high La/Zr) signatures. Bulk mixing between an enriched mantle (EM) source and metasedimentary lower crustal material (MLC) (composition derived from Harangi *et al.*, 2001) would suggest that addition of 5-10% of MLC would produce the isotopic compositions of Apuseni magmas (Fig. 7). Data for sample 790 (representing the oldest magmatism) plot away from the main group of samples, which may suggest that both AFC and source contamination operated at the onset of the magmatic activity.

The Apuseni and South Harghita magmas show a similar restricted range of low  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.7045-0.706) that may suggest a depleted source for both areas, but with much lower  $\delta^{18}\text{O}$  values for the Apuseni Mountains. The shift to higher  $\delta^{18}\text{O}$  values in South Harghita, combined with the small variation in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, has been considered by Peccerillo and Taylor (1976) and Mason *et al.* (1996) as related to contamination in the lower crust rather than the upper crust. According to Seghedi *et al.* (2004), the South Harghita magmas formed by mixing between slab-derived melts with relatively high  $\delta^{18}\text{O}$  values (6-6.9 ‰) and melts from a variably enriched mantle source.

### ***Source of Apuseni Mountains magmas***

The real nature of the source of the Apuseni Mountains magmas is difficult to envision. However, the presence of garnet-bearing andesites among the earliest volcanic rocks in the area leads us to the assumption that the source could be the lower crust that may include mafic garnet granulites. Such granulites are found as xenoliths brought to the surface by alkali basaltic magmas in the neighbouring Pannonian Basin (Kempton *et al.*, 1997, Dobosi *et al.*, 2003 and Embey-Isztin *et al.*, 2003), although they represent the lower crust of the Alcapa terrane rather than the Tisia-Dacia block. They are isotopically and lithologically heterogeneous. Along with mafic granulite xenoliths, metasedimentary lower crustal material has been found, whose isotopic composition was used in this paper as the possible lower crustal component for mixing in our modelling (Fig. 7).

Drummond *et al.* (1996) reported the augmentation of Sr content in adakitic melts formed by high-pressure partial melting of basalts, where it behaves incompatibly due to the absence or instability of plagioclase, whereas Y is controlled by the presence of garnet in the residuum. A plot of  $\delta^{18}\text{O}$  vs. Sr/Y (Defant and Drummond, 1990) was used to differentiate between adakite-type magmas and typical calc-alkaline magmas (Fig. 8). The diagram indicates two contrasting trends for calc-alkaline magmatism in the Carpathian-Pannonian region: a) low Sr/Y ratios with large  $\delta^{18}\text{O}$  variation for volcanic rocks from the Călimani and Central Segment volcanic areas; b) large Sr/Y variation with a narrow range of  $\delta^{18}\text{O}$  variation for magmas from the Apuseni and South Harghita areas. In these latter regions the magmas show an adakite-like character; in the Apuseni Mountains this is seen in most of the rocks younger than 12 Ma. They display a narrow range of  $\delta^{18}\text{O}$  variation, correlating with low  $^{86}\text{Sr}/^{87}\text{Sr}$  ratios (c.a. 0.7045), with higher  $\text{K}_2\text{O}$  and Rb and relatively high in Mg#, Cr and Ni contents (Mason *et al.*, 1996; Roşu *et al.*, 2004a). These rocks show high Mg#s in most of their clinopyroxenes, which suggests that the magmas may represent primary melts (Bindeman *et al.*, 2005), whereas the absence of olivine phenocrysts may be interpreted to indicate an olivine-free source. Most adakites, e.g. from Adak and Cook Islands, also lack olivine and contain only clinopyroxene phenocrysts with similar  $\delta^{18}\text{O}$  values to those from the Apuseni samples (Bindeman *et al.*, 2005). On the other hand a narrow range and low  $\delta^{18}\text{O}$  variations in pyroxene and amphibole in post-12 Ma Apuseni Mountains magmas that show variable Sr/Y ratios suggest a magma source that was homogeneous in terms of  $\delta^{18}\text{O}$ , but compositionally heterogeneous. This supports the interpretation that melting took place at a deep level in the lower crust and/or in the enriched lithospheric mantle (Roşu *et al.*, 2001, 2004a; Seghedi *et al.*, 2004). Also the hydrogen isotope composition shows a trend with time, the younger rocks having lower  $\delta\text{D}$  values, suggesting a change in source composition.

We have used a plot of  $\delta^{18}\text{O}$  mineral compositions vs. Nb/Zr in the whole-rocks (Fig. 9), since this ratio gives constraints on different mantle source components varying from  $\sim 0.03$  (MORB value) up to 0.20 (OIB values). Different Nb/Zr ratios are generally interpreted in terms of variations in source composition and/or changes in degree of partial melting of the mantle (e.g. Thirlwall *et al.*, 1994; Singer *et al.*, 1996). Samples

from the subduction-related Călimani and South Harghita volcanic areas in the Carpathian arc show a narrow range of Nb/Zr, suggesting that they were derived from a chemically rather homogeneous source. Data for the Central Segment volcanic area show large variations in both  $\delta^{18}\text{O}$  and Nb/Zr ratios that are spatially controlled (transversely to the collision front of the Carpathians; Nb/Zr values are lower in magmas formed nearer to the inferred trench) (Seghedi *et al.*, 2001). The Apuseni Mountains magmas show a similar variation in Nb/Zr ratios as subduction-related magmas from elsewhere in the Carpathian-Pannonian region, but very little variation in  $\delta^{18}\text{O}$ . This implies either source variation or variable degrees of melting of a source that was homogeneous with respect to its oxygen isotopic composition. This is different from typical adakite rocks that show source characteristics closer to MORB (Defant *et al.*, 2002). The  $\delta^{18}\text{O}$  values of Apuseni magmas are slightly higher than those of the typical local OIB-type source, represented by olivine phenocrysts from alkali basalts of the Carpathian-Pannonian region (Dobosi *et al.*, 1998). This supports the suggestion of chemically heterogeneous, but isotopically uniform sources for magmas younger than 12.5 Ma, probably of lower crustal and/or lithospheric mantle origin (Roşu *et al.*, 2004a).

### ***Model for magmagenesis in the Apuseni Mountains***

Ever since the term “adakite” was introduced by Defant and Drummond (1990), debate has emphasized that the term should not be restricted to processes related to slab melting, but can also be applied to those involving melting of the lower crust (e.g. Defant *et al.*, 2002; Hou *et al.*, 2004; Wang *et al.*, 2006). Generation of magmas with unusual geochemical characteristics (e.g. high Sr, Sr/Y and La/Yb values) can occur by the following mechanisms: a) partial melting of subducting oceanic crust at depths where garnet is stable in the residue (e.g. Kay *et al.*, 1993; Gutscher *et al.*, 2000; Sajona *et al.*, 2000); b) partial melting of thickened lower continental crust (e.g. Atherton and Petford, 1993; Wang *et al.*, 2005) or c) partial melting of delaminated mafic lower continental crust that converts to eclogite (e.g. Kay and Kay, 1993; Defant *et al.*, 2002; Xu *et al.*, 2002; Gao *et al.*, 2004).

Tectonic reconstructions (Fig. 2) and the seismic reflection profiles along the western margin of the Apuseni Mountains (Fig. 1) indicate an important rise of the lithosphere-asthenosphere boundary beneath the Zarand basin. This geodynamic situation is the result of extension and suggests that magma generation in this region cannot be associated with contemporaneous subduction, but instead is related to fast rotational tectonics in Miocene times (e.g. Seghedi *et al.*, 2004; Roşu *et al.*, 2004a). The isotopic data presented in this paper support the conclusion that the Apuseni Mountains magmatism was not related to subduction, but instead resulted from an unusual extensional tectonic setting. As discussed above, recent hypotheses imply adakite-type magma generation may result from lower crustal melting. Two mechanisms have been invoked. The first is differentiation and/ or partial melting near the base of the subcontinental arc crust by re-melting ponded basalts and/or mafic cumulates at depths of at least 32–35 km (e.g. Bindeman *et al.*, 2005). This mechanism is probably not viable for the Apuseni Mountains magmatic province, where the present-day base of the crust is only at ~ 30 km (Takács *et al.*, 1996, Rădulescu and Diaconescu, 1998). An alternative hypothesis is that mafic lower crust might convert to eclogite and partially melt even in convergent margins with thin crust (<30 km), if it delaminates and sinks into the underlying mantle (e.g. Defant *et al.*, 2002). Such a mechanism is possible in the Apuseni case and is illustrated on the lithospheric cross-section shown in Fig. 1. We favor a close relationship between tectonic processes and magma generation as follows:

1. The period between 15 and 13.5 Ma was a time of fast rotations and the onset of magma generation. The resulting rocks show relatively high  $^{86}\text{Sr}/^{87}\text{Sr}$  ratios, the highest amphibole  $\delta^{18}\text{O}$  and the lowest groundmass  $\delta\text{D}$  values and are probably a consequence of crustal decompression melting during this initial phase of extension. The observed isotope heterogeneity of lower crustal granulite xenoliths from the Pannonian Basin (Kempton *et al.*, 1997; Dobosi *et al.*, 2003) may explain the variable  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  values of the source of the early magmatism.

2. Between 13.5 and 12.5 Ma, the amount of rotation diminished. This was the time-interval during which normal calc-alkaline magmas were generated, with lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  values than the older ones. This was probably a period of increasing temperature and pressure at the base of the crust due to rotational shear heating, which

started to convert mafic lower crust into amphibole-bearing eclogites and produced normal calc-alkaline magmas, including garnet-bearing ones (unpublished data).

3. In the time-interval 12.5-10 Ma, after rotation ended, mainly adakite-like, but also normal calc-alkaline magmas were generated, which have lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  values in their early magmatic minerals. This was the main period of extension, volcanism and metallogenesis in the Apuseni Mountains (Roşu *et al.*, 2004b). The province contains one of the largest gold ore deposits in Europe and adakites are known to be linked to epithermal gold deposits (e.g. Thieblemont *et al.*, 1997; Defant *et al.*, 2002; Wang *et al.*, 2005). We suggest that formation of high density garnet-bearing (eclogitic) lower crust led to delamination, followed by partial melting and generation of adakite-like magmas in the underlying mantle.

4. From 10 to 7 Ma a limited volume of magma was erupted. It still retained a slight adakitic character, as well as lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  mineral values, but shows an increased Nb/Y ratio, suggesting upwelling of hotter asthenospheric mantle and mixing processes between melts from the delaminated crust and those from the asthenosphere.

5. After a major time gap, the final volcanic episode at 1.6 Ma was the eruption of a small volume of alkali trachyandesite magmas at Uroi Hill, which also show low  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  pyroxene values. These rocks are characteristically anhydrous as indicated by the absence of water in the groundmass and the presence of fluor-amphiboles (Bojar and Walter, 2006). Seghedi *et al.* (2004) related this episode to recent extension that reactivated already upwelled asthenospheric mantle along deep fault zones; mantle asthenosphere partial melts mixed with the lithospheric mantle melts, triggered by the new influx of heat from the asthenosphere.

### ***Summary and Conclusions***

Volcanic rocks from the Apuseni Mountains Neogene magmatic province have  $\delta^{18}\text{O}$  values of 5.5-5.9 ‰ for clinopyroxenes, biotites and amphiboles, slightly higher than those of phenocrysts in alkali basalts from elsewhere in the Carpathian-Pannonian region (5.0 - 5.34 ‰, Dobosi *et al.*, 1998). The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for clinopyroxenes (~0.7041) and amphiboles (0.7042 - 0.7042) are lower than whole rock  $^{87}\text{Sr}/^{86}\text{Sr}$ , but values

for the plagioclase and groundmass are generally higher. The Mg-rich nature of the pyroxene phenocrysts suggests that they may have crystallized from primary melts. The higher  $\delta^{18}\text{O}$ ,  $\delta\text{D}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  for the oldest rocks (15-13.5 Ma) suggest a different enriched (possibly lower crustal) source. Rocks younger than 13.5 Ma have uniformly lower  $\delta^{18}\text{O}$ ,  $\delta\text{D}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  in their mafic phenocrysts, which correlate with the narrow range and low values of whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, different from most other calc-alkaline volcanic rocks in the Carpathian-Pannonian region. This suggests a source that had a rather uniform  $\delta^{18}\text{O}$  composition with chemical heterogeneities, and enables us to reject the hypothesis of a mantle-source influenced by subduction-related metasomatism. On the other hand our study demonstrates the importance of fractional crystallization, with a small amount of assimilation for some of the rocks that show an increase in  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in their later phases.

It is plausible that three melting mechanisms may have operated individually or coupled (leading to local magma mixing) to generate the complex calc-alkaline non-subductional magmatic association in the Apuseni Mountains. Most probably the order of events was as follows: (1) lower crustal decompression melting as a result of extensional tectonics (calc-alkaline magma generation between 14.5 and 13.5 Ma); (2) lower crustal delamination and eclogite generation by terrane rotation and lower crustal shear heating (normal calc-alkaline and mainly adakite-like magma generation between 13.5 and 7 Ma) and (3) asthenospheric upwelling triggering small degree melting, followed by magma mixing within the lithospheric mantle (alkaline magma generation, at 1.6 Ma).

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### **Figure and table captions**

Table 1. Location, lithology and whole rock compositions of selected rocks from the Apuseni Mountains, analysed in this study. Age determinations (K/Ar method) are from Pécskay *et al.* (1995) and Roşu *et al.* (1997, 2004b) and whole rock geochemistry from Roşu *et al.* (2004a).

Table 2. Chemical composition of cores (c) and mantles (m) of clinopyroxenes from selected Apuseni Mountains Neogene magmatic rocks.

Table 3. Oxygen, Hydrogen, water content and Sr isotope composition of minerals and groundmass from Apuseni Mountains Neogene magmatic rocks. Abbreviations: q-quartz; am-amphibole; bi-biotite; px-pyroxene

Fig 1. Simplified sketch of Apuseni Mountains of western Romania showing the Neogene magmatic area (dark grey) and sample collection points. Inset shows the geographical setting and location of the PGP-4 profile. Integrated interpretation of the low-frequency seismic reflection PGP-4 profile (Takács *et al.*, 1996) indicating an important asthenosphere rise along lithosphere thinning below the Bekes and Zarand basins. Tectonic interpretation of the profile on the Hungarian territory is after Csontos *et al.*, 2002 and on the Romanian territory is according to Săndulescu, 1988. Magma generation processes are suggested for: (1) calc-alkaline magma generation by decompressional crustal melting at 14.5-13.5 Ma and (2) adakite-like magma generation by lower crustal delamination and eclogite formation during terrane rotation between 13.5-7 Ma.

Fig. 2. Two schematic examples of geodynamic evolution in Carpathian-Pannonian region during Middle to Late Miocene times. The right-hand figure is from Csontos *et al.* (2002), showing the geodynamic situation after the end of major rotations in the east (Pannonian time, at ~12-11 Ma). Differential rotation of the central (black arrow) and eastern parts (white arrow) of Tisia can be noted, as well as the graben opening in the western part of Apuseni Mountains. The left-hand figure is from Seghedi *et al.* (2004) for the 15.5 to 11 Ma time-interval, showing that magmatism in the Apuseni Mountains is closely connected with the major eastward rotations and subsequent graben-type basin opening. Active and already extinct volcanoes are simplified. Clockwise rotation around a pole situated in western Moesia (Csontos and Nagymarosy, 1998) facilitated thinning of the lithosphere and *en-echelon* brittle crustal splitting.

Fig. 3 (a) Nb/Y and (b) Sr/Y vs. age for Neogene magmatic rocks from the Apuseni Mountains (triangles), highlighting the samples used in this study (squares). Data are from Roşu *et al.*, 2004a. Shaded areas represent 70° and 30° angle of declination suggesting main Miocene clockwise rotations in the Apuseni Mountains area (Panaiotu, 1998, 1999; Roşu *et al.*, 2004a).

Fig. 4 a.  $\delta^{18}\text{O}$  value of minerals and groundmass vs. whole-rock  $\text{SiO}_2$ ; b.  $\delta^{18}\text{O}$  value of minerals vs. whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$ ; c. Mineral  $^{87}\text{Sr}/^{86}\text{Sr}$  vs. mineral  $\delta^{18}\text{O}$  compositions for

mineral separates and groundmass in selected Neogene magmatic rocks from the Apuseni Mountains Stars represent the SiO<sub>2</sub> mantle composition and the whole rock <sup>87</sup>Sr/<sup>86</sup>Sr ratios for depleted and enriched mantle in CPR, calculated from the composition of alkali basalts, as compiled by Mason *et al.* (1996).

Fig. 5 a. δD of mineral phases and groundmasses vs. whole-rock SiO<sub>2</sub> in selected Neogene magmatic rocks from the Apuseni Mountains.; b. δD of mineral phases and groundmasses versus age.

Fig. 6. δ<sup>18</sup>O value of minerals vs. SiO<sub>2</sub> whole-rock values of several Neogene volcanic regions in the Carpathian-Pannonian region. Abbreviations: C-Călimani area; CS- Central segment volcanic area (as defined by Seghedi *et al.*, 2004); Ap-Apuseni; SH-South Harghita volcanic area; AB- local alkali-basalts; FC-fractional crystallization; AFC-assimilation-fractional crystallization. Data from Downes *et al.*, 1995; Mason *et al.*, 1996; Dobosi *et al.*, 1998; Seghedi *et al.*, 2001, 2004.

Fig. 7. δ<sup>18</sup>O value of minerals vs. <sup>87</sup>Sr/<sup>86</sup>Sr ratios for several Neogene volcanic regions in the Carpathian-Pannonian region (<sup>87</sup>Sr/<sup>86</sup>Sr values for Apuseni Mts are mafic mineral values, whereas those for the other regions are whole-rock values). Abbreviations: C-Călimani area; CS- Central segment volcanic area (acc. Seghedi *et al.*, 2004); Ap-Apuseni; SH-South Harghita volcanic area; AB- local alkali-basalts; AFC-assimilation-fractional crystallization. Data from Downes *et al.*, 1995; Mason *et al.*, 1996; Dobosi *et al.*, 1998; Seghedi *et al.*, 2001, 2004. Mixing lines have been constructed between possible mantle melts (derived from local depleted mantle-[DM]- and local enriched mantle-[EM]- using data from Embey-Isztin *et al.*, 1993; Downes *et al.*, 1995) and sediment end-members (average value for East Carpathians local sediments from Mason *et al.*, 1996). O-Sr isotopic modeling of bulk mixing between these mantle sources and sediments produces mixing curves which indicate source contamination in the range of 0.5-1% for samples from the Apuseni and 1-3 % for Călimani and CS volcanic areas using DM or 1-3 % and 3-5 % for using EM. Bulk mixing between the EM source and

metasedimentary lower crustal material (MLC) (composition derived from Harangi *et al.*, 2001) would suggest that 3-7 % mixing is needed to produce the isotopic compositions of Apuseni magmas. Assimilation-fractional crystallisation curves (AFC) have been modelled using the most isotopically primitive compositions from the Călimani volcanic structure (sample C65) and CS (sample 134t) and a crustal assimilant (average value for East Carpathians local crust from Mason *et al.*, 1996). Between 5 and 20 % upper crustal contaminant is required in the AFC modelling for these areas (Seghedi *et al.*, 2004). Despite the lack of a proper primitive composition for the Apuseni mountains samples, if we apply AFC modelling, < 3% assimilation can account for the observed range.

Fig. 8.  $\delta^{18}\text{O}$  values of minerals vs. Sr/Y whole-rock compositions of several Neogene volcanic regions in the Carpathian-Pannonian region. Abbreviations: C-Călimani area; CS- Central segment volcanic area (defined by Seghedi *et al.*, 2004); Ap-Apuseni; SH- South Harghita volcanic area; AB- local alkali-basalts. Data from Table 3 and Downes *et al.*, 1995; Mason *et al.*, 1996; Dobosi *et al.*, 1998; Seghedi *et al.*, 2001, 2004.

Fig. 9.  $\delta^{18}\text{O}$  values of minerals vs. Nb/Zr whole-rock compositions of several Neogene volcanic regions in the Carpathian-Pannonian region. Abbreviations: C-Călimani area; CS- Central segment volcanic area (defined by Seghedi *et al.*, 2004); Ap-Apuseni; SH- South Harghita volcanic area; AB- local alkali-basalts. Data from Table 3 and Downes *et al.*, 1995; Mason *et al.*, 1996; Dobosi *et al.*, 1998; Seghedi *et al.*, 2001, 2004.

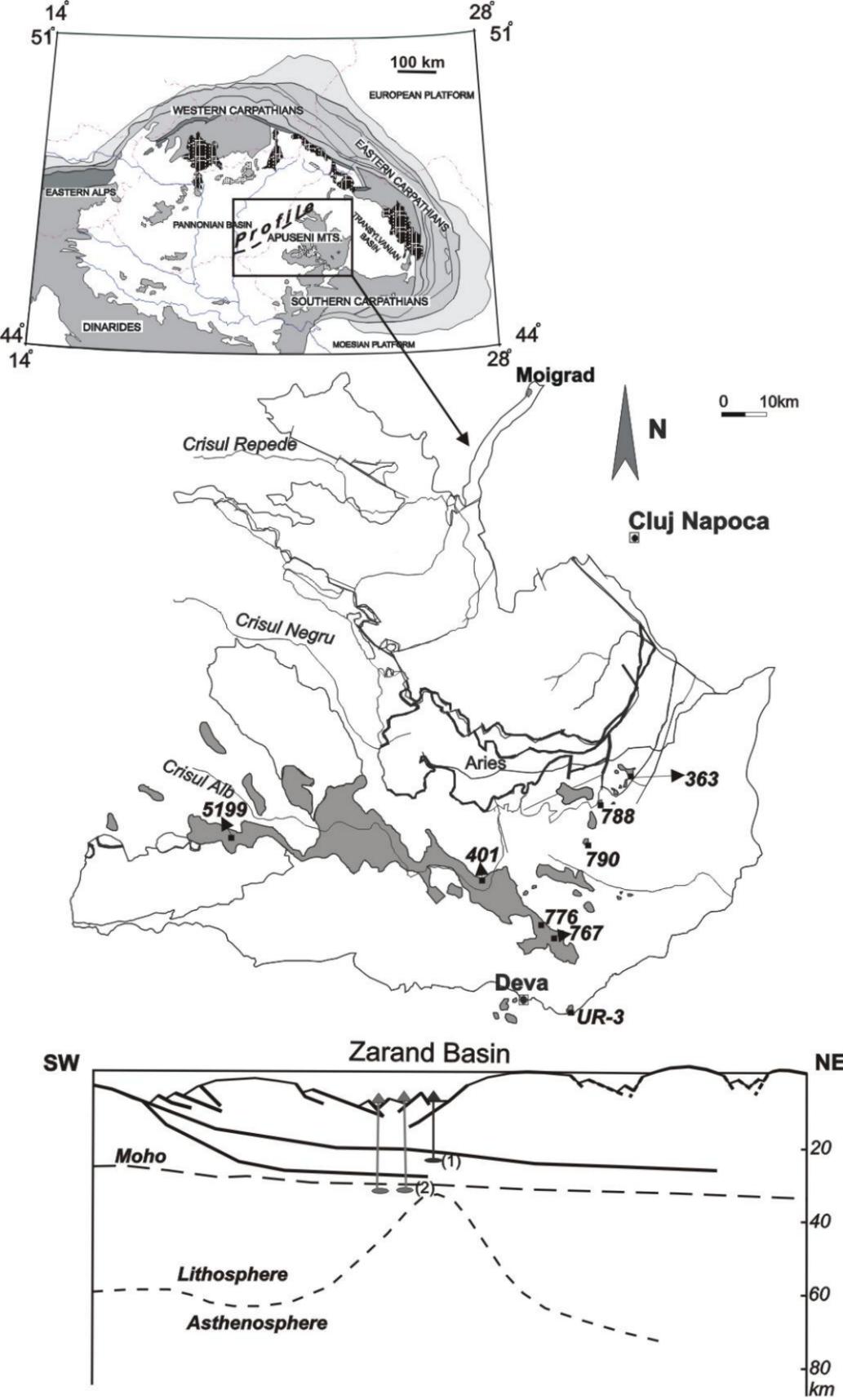
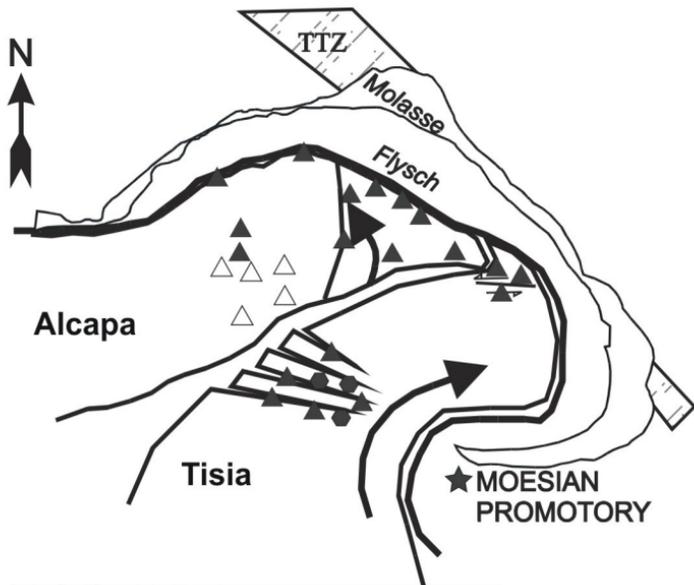


Fig. 1

# Seghedi et al., 2004



<b>VOLCANISM</b>	Active	▲	Calc-alkaline
	Extinct	△	
		●	Adakitic Calc-alkaline & Alkaline

# Csontos et al., 2002

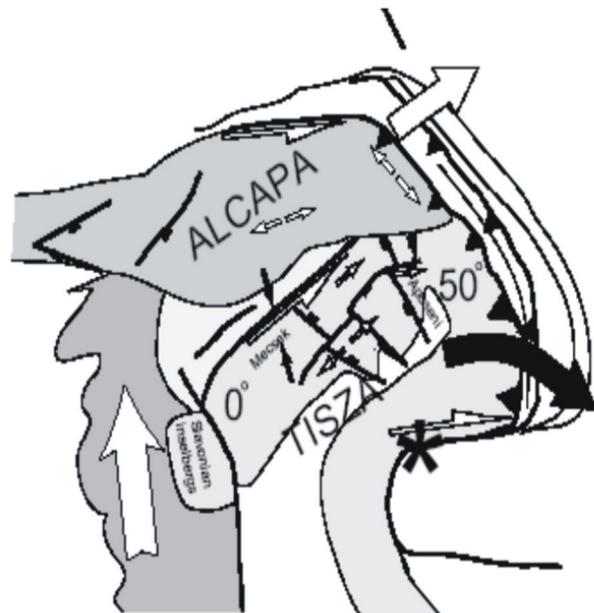
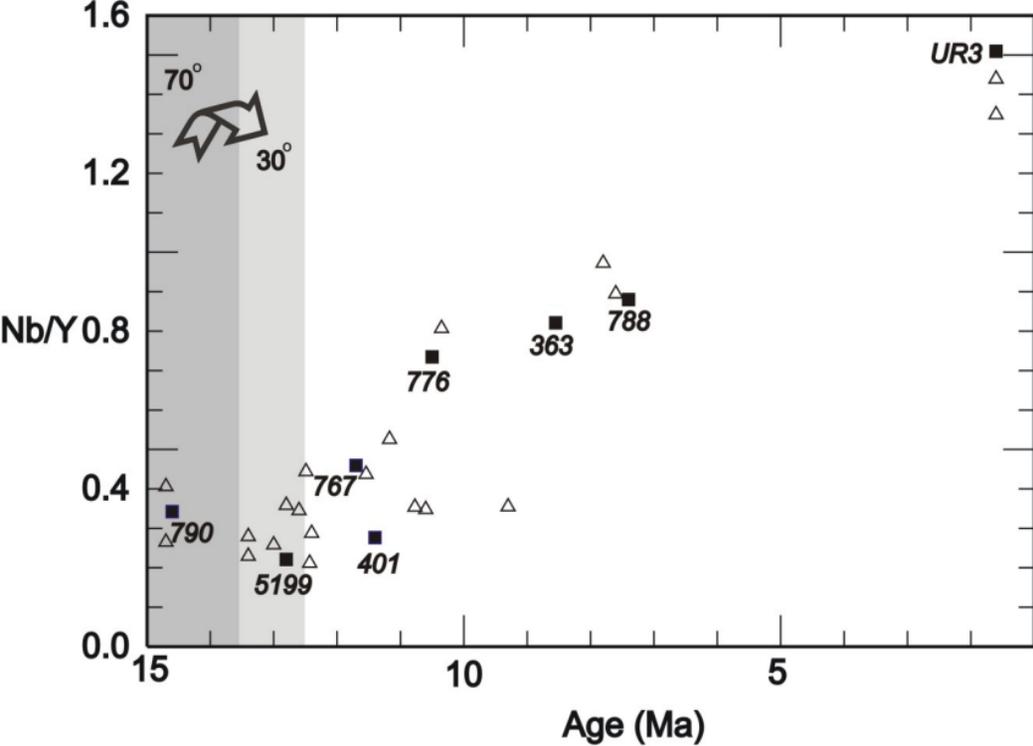
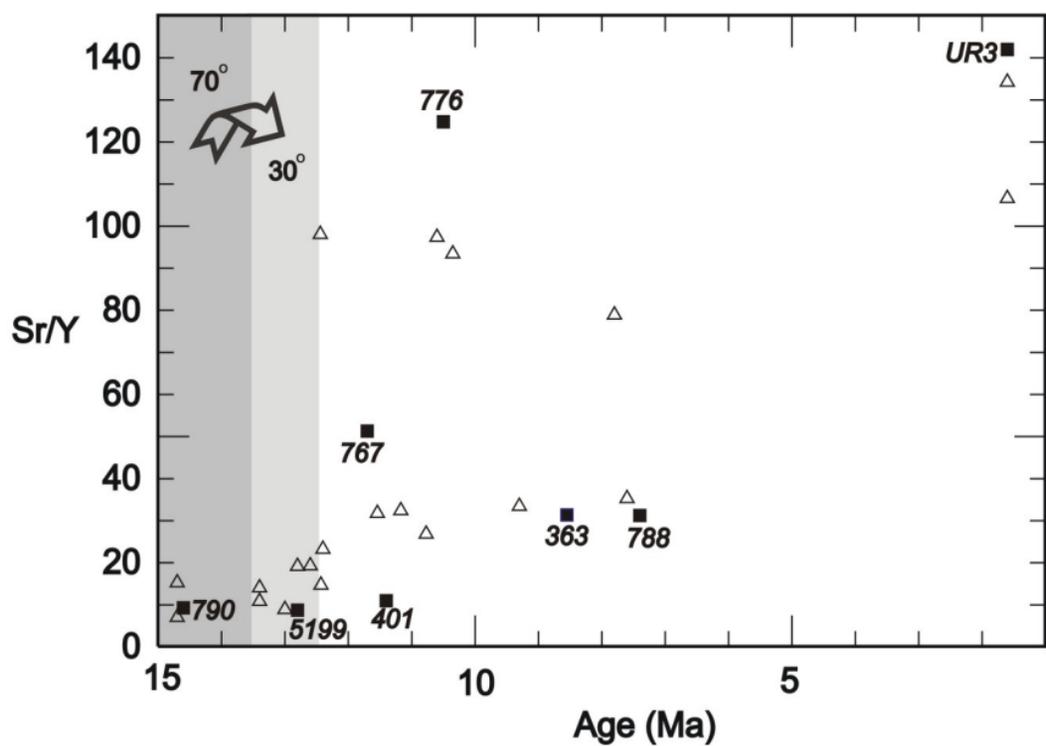


Fig. 2



a.



b.

Fig. 3

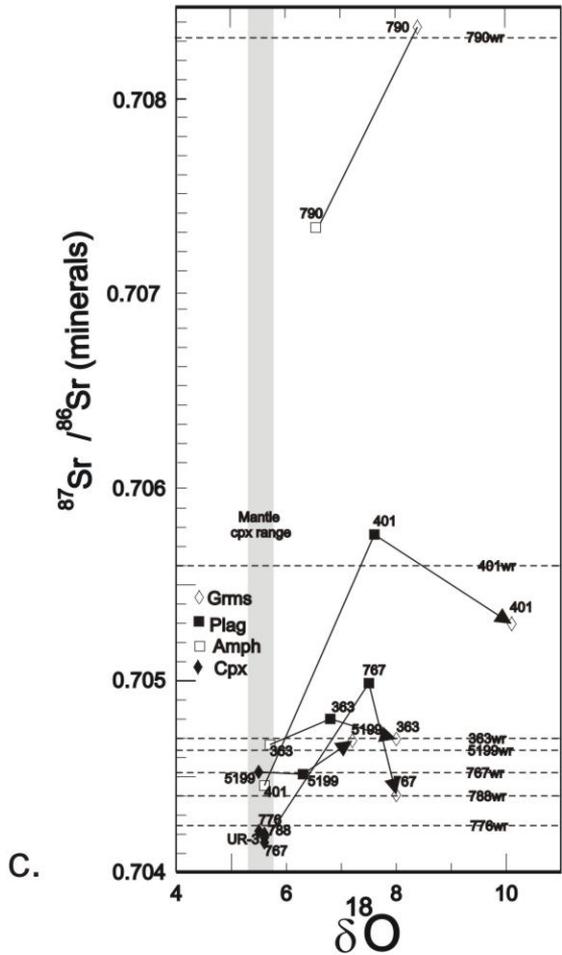
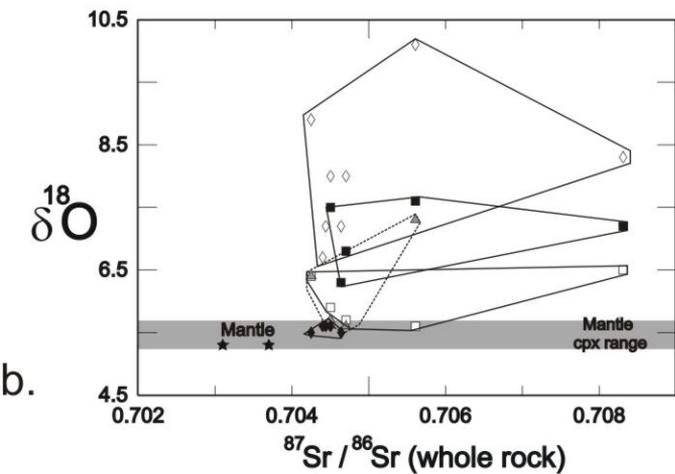
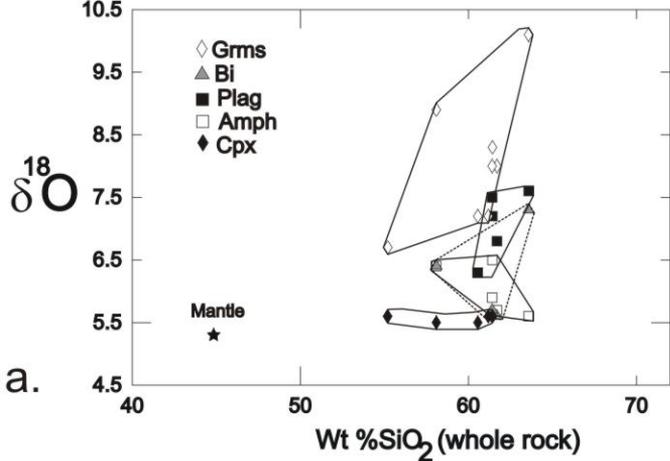


Fig. 4

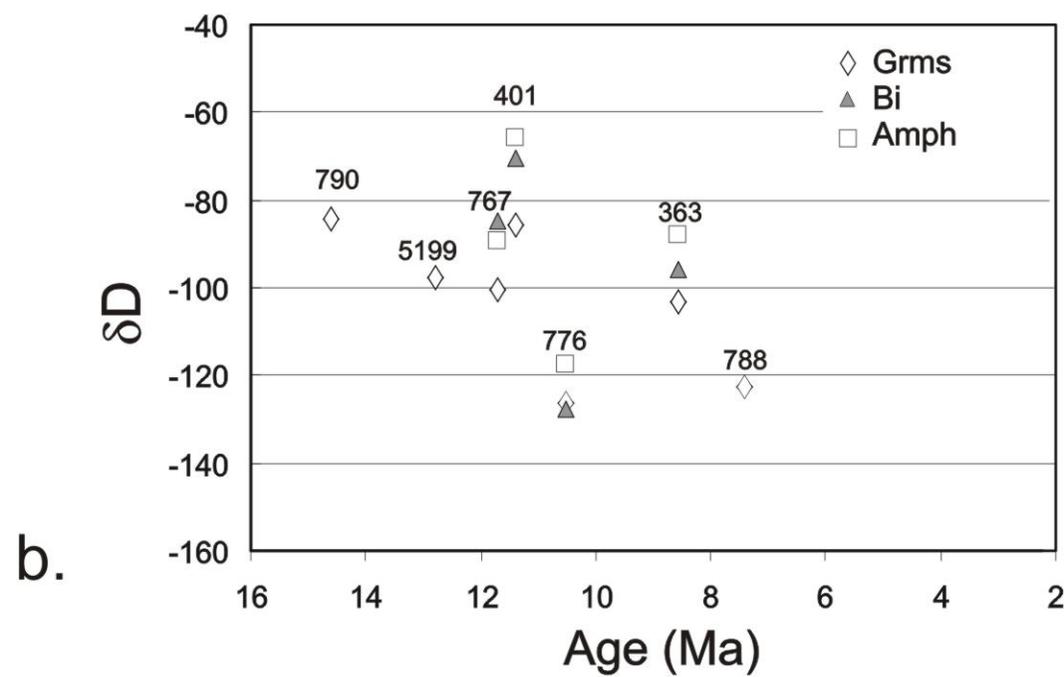
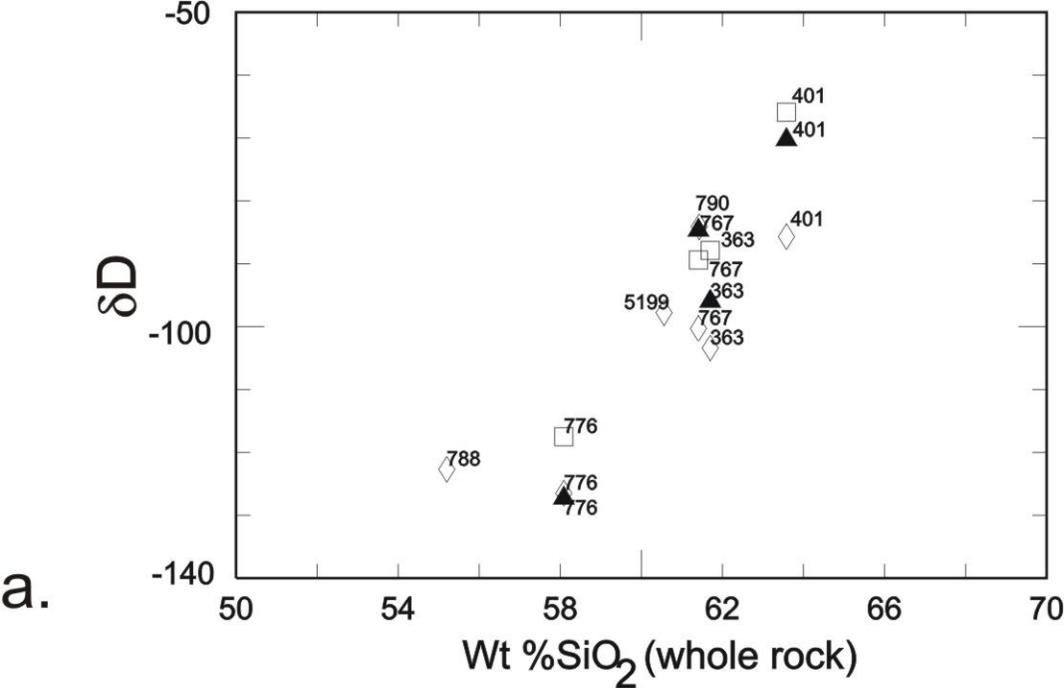


Fig. 5

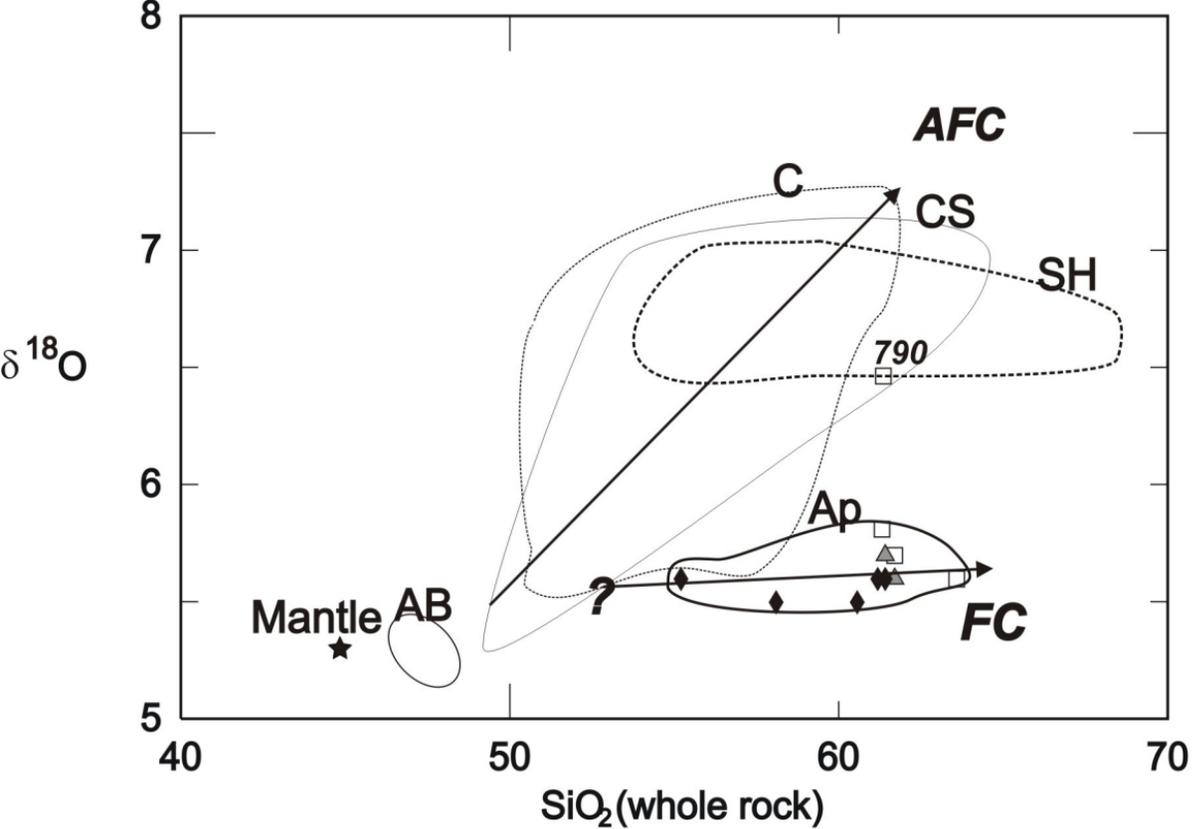


Fig. 6

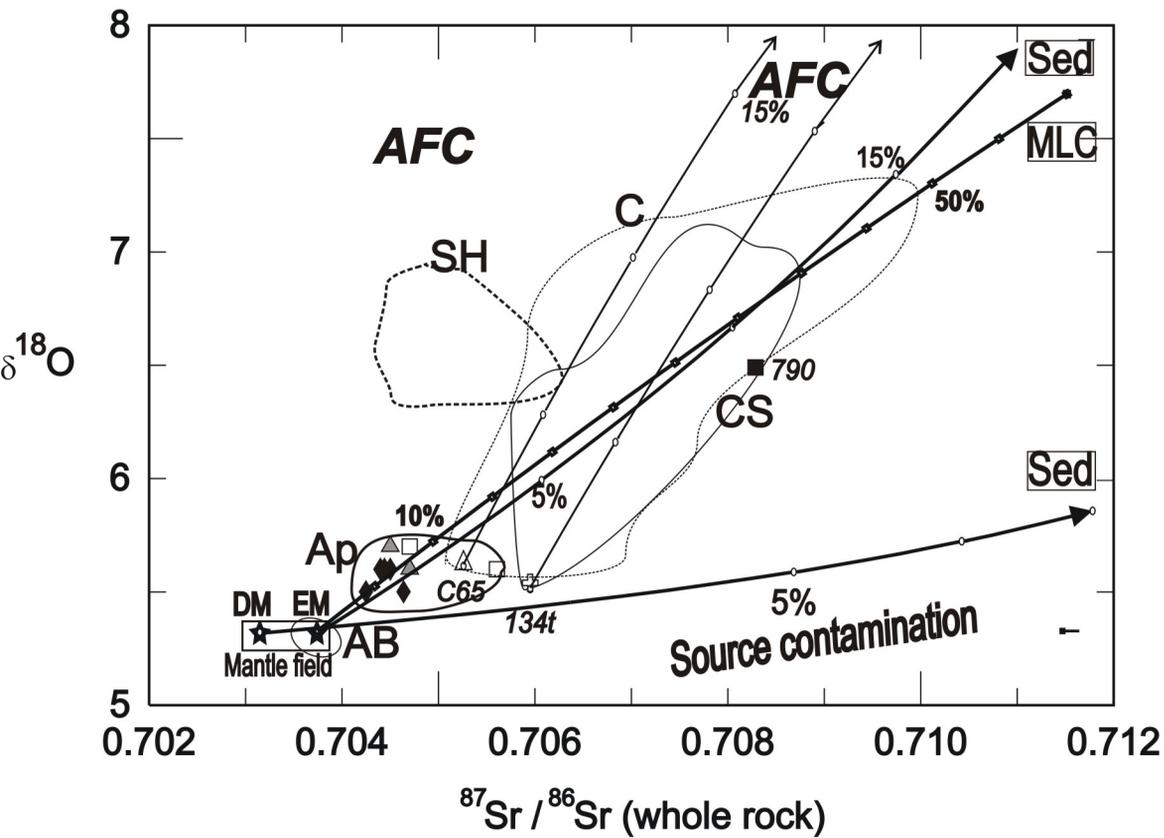


Fig.7

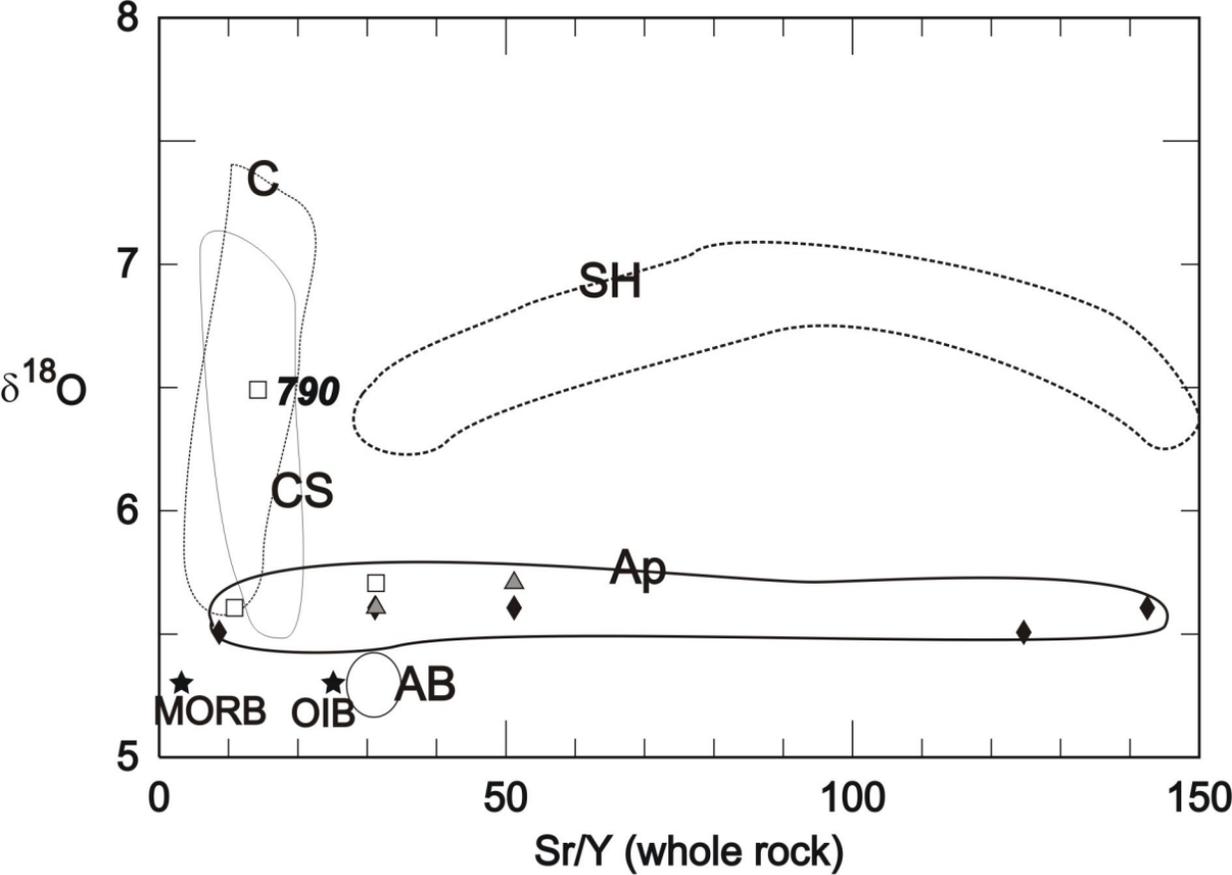


Fig.8

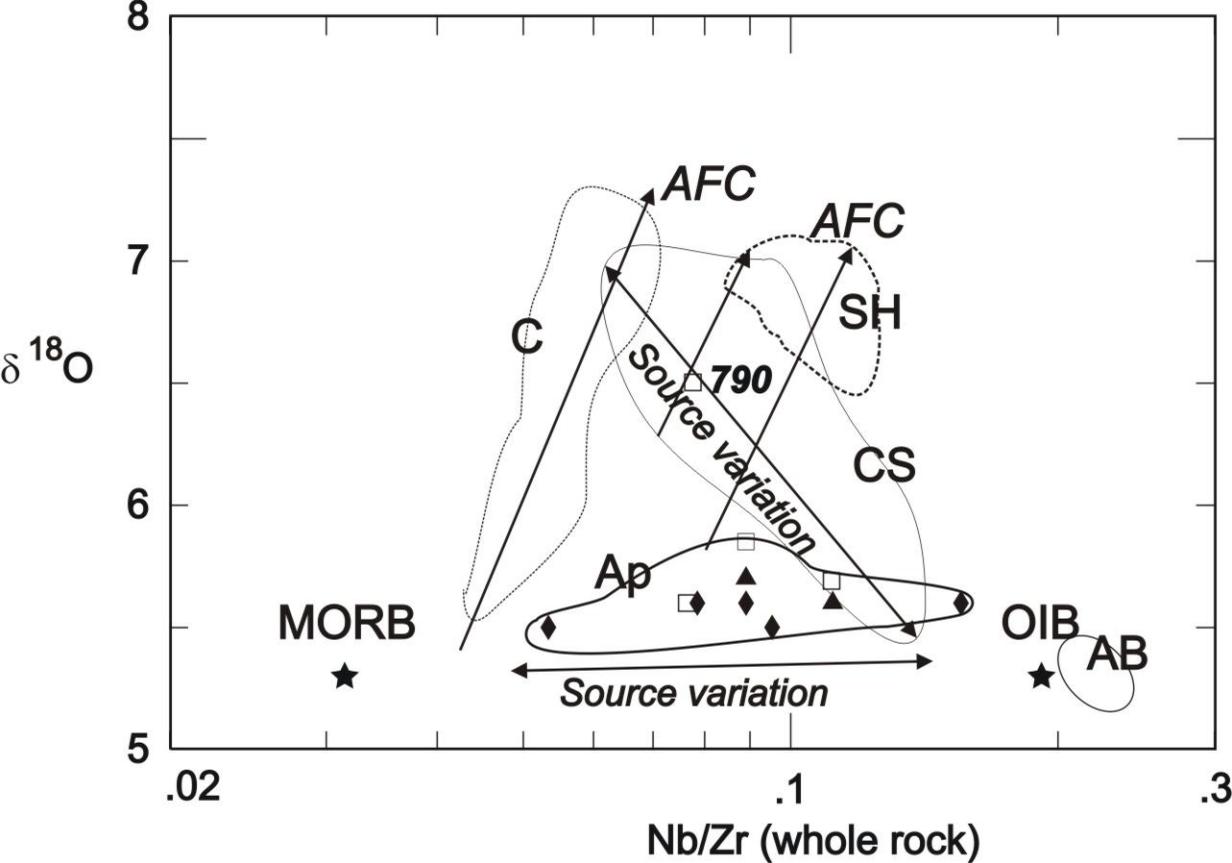


Fig. 9

Table 1. Location, lithology and whole rock compositions of selected rocks from the Apuseni Mountains, analyzed in this study, Age determinations (K/Ar method) and geochemistry are from Pécskay et al., 1995 and Roşu et al., 1997, 2004

Sample	Locality	Lat.	Long.	SiO <sub>2</sub> (%)	K <sub>2</sub> O (%)	Sr (ppm)	Y (ppm)	Nb (ppm)	Zr (ppm)	Age (Ma)	<sup>87</sup> Sr/ <sup>86</sup> Sr (wr)	<sup>143</sup> Nd/ <sup>144</sup> Nd (wr)
790	Citera	46.233	23.213	61.43	1.39	212	22.8	7.8	101	14.6	0.708312	0.512421
5199	Chisindia	46.278	22.081	60.56	1.30	249	28.5	6.3	118	12.8	0.704640	0.512778
767	Cetras	46.033	22.973	61.41	1.47	1005	19.6	9	101	11.7	0.704502	0.512634
401	Brad	46.138	22.844	63.58	1.89	321	29.3	8.1	106	11.4	0.705608	0.512593
776	Zambrita	46.045	22.953	58.09	2.92	2770	22.2	16.3	171	10.5	0.704250	0.512609
363	Poienita	46.326	23.286	61.70	1.99	665	21.2	17.4	156	8.55	0.704704	0.512667
788	Detunata	46.278	23.199	55.20	1.45	596	19.1	16.8	108	7.4	0.704400	0.512689
UR-3	Uroi	45.857	23.044	61.18	5.32	2353	16.5	24.9	317	1.6	0.704441	0.512680

Table 2  
Chemical composition of clinopyroxenes from Apuseni rocks

Sample	5199/ 214	5199/ 210	767/ 45	767/ 40c	767/ 40m	776/ 205	776/ 206	788/ 60	788/ 50c	788/ 50m	UR-3/ 236	UR-3/ 224c	UR-3/ 224m
SiO <sub>2</sub>	52.57	51.51	53.09	50.85	53.42	53.45	53.19	53.53	53.63	51.43	53.78	51.94	52.84
TiO <sub>2</sub>	0.27	0.37	0.17	0.35	0.14	0.15	0.20	0.31	0.25	0.85	0.29	0.07	0.41
Al <sub>2</sub> O <sub>3</sub>	2.04	2.30	1.63	2.90	1.15	1.43	1.53	1.46	1.37	1.80	1.35	1.16	1.75
Cr <sub>2</sub> O <sub>3</sub>	0.21	0.17	0.68	0.10	0.20	0.75	0.29	0.42	0.75	0.07	0.01	0.05	0.03
FeO	7.03	7.96	3.32	9.57	3.90	3.16	3.55	3.07	2.96	9.42	4.34	11.31	4.98
MnO	0.17	0.22	0.11	0.26	0.12	0.10	0.09	0.06	0.07	0.31	0.12	0.21	0.13
MgO	15.98	15.90	17.64	12.16	18.01	17.96	17.35	17.60	18.02	15.60	16.66	10.73	16.00
CaO	20.55	19.70	22.28	21.50	21.66	21.84	22.47	22.72	22.29	18.74	21.70	22.88	21.64
Na <sub>2</sub> O	0.22	0.26	0.27	0.80	0.16	0.24	0.23	0.22	0.24	0.34	0.51	0.37	0.50
Total	99.03	98.39	99.19	98.49	98.76	99.08	98.89	99.39	99.59	98.57	98.74	98.72	98.28
Wo	38.92	36.52	41.14	40.50	41.1	40.95	42.31	42.74	41.57	35.65	42.34	46.37	42.00
En	44.25	44.38	48.13	34.34	49.36	49.02	47.53	47.94	48.94	43.71	45.81	30.68	44.36
Fs	11.19	11.18	3.67	12.97	5.25	4.51	4.62	4.26	3.93	13.95	6.88	18.49	7.76
Mg#	80.21	78.08	90.45	69.37	89.18	91.01	89.71	91.10	91.55	74.69	87.26	62.83	85.13

Table 3. Oxygen, Hydrogen, water content and Sr isotope composition of minerals and groundmass from Apuseni Mountains Neogene magmatic rocks.

Sample	Mineral	$\delta^{18}\text{O}\text{‰}$ SMOW	dD $\text{‰}$ SMOW	Water content (wt%)	$^{87}\text{Sr}/^{86}\text{Sr}$ (min)
790	Amph	6.5			0.707416 $\pm$ 15
	Pl	7.2			0.708147 $\pm$ 8
	Gm	8.3	-84.1	2.2	0.708380 $\pm$ 9
5199	Cpx	5.5			0.704528 $\pm$ 9
	Pl	6.3			0.704513 $\pm$ 17
	Gm	7.2	-97.8	1.1	0.704688 $\pm$ 9
767	Cpx	5.6			0.704157 $\pm$ 11
	Amph	5.9	-89.4	2.4	
	Bi	5.7	-84.7	2	
	Pl	7.5			0.704987 $\pm$ 9
	Gm	8	-100.3	1.4	0.704405 $\pm$ 9
401	Amph	5.6	-65.9	2.2	0.704454 $\pm$ 10
	Bi	7.3	-70.3	4.4	
	Pl	7.6			0.705762 $\pm$ 9
	Gm	10.1	-85.7	2.4	0.705296 $\pm$ 9
776	Cpx	5.5			0.704216 $\pm$ 11
	Amph	6.4	-117.5	2.3	
	Bi	6.4	-127.5	2.3	
	Gm	8.9	-126.5	1.5	
363	Amph	5.7	-87.9	2.4	0.704667 $\pm$ 10
	Bi	5.6	-96.0	3.6	
	Pl	6.8			0.704801 $\pm$ 8
	Gm	8	-103.4	3.6	0.704696 $\pm$ 11
788	Cpx	5.6			0.704202 $\pm$ 8
	Gm	6.7	-122.7	0.8	
UR-3	Cpx	5.6			0.704183 $\pm$ 13
	Gm	7.2			