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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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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. Magmatic rocks older than 13.5 Ma are characterised by the highest $^{87}Sr/^{86}Sr$, $\delta^{18}O$ and δD 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 $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ ratios, with a narrow range for pyroxene and amphibole mineral separates, indicating closed system fractional crystallization. A larger scatter of $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ values is found in plagioclase phenocrysts and groundmasses, indicating that a small amount of crustal assimilation also occurred. The δD values for hydrogen-bearing phases such as amphibole, biotite and groundmass indicate fractional crystallisation in a closed system and a trend of decreasing δD 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 mafic lower 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.

Keywords: Carpathian–Pannonian region; Apuseni Mountains; Magmagenesis; $^{87}Sr/^{86}Sr$; Stable isotope geochemistry

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1. 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). Whereas the main Carpathian volcanic arc is related to subduction (Seghedi et al., 2004; Harangi et al., 2006), the Apuseni province is situated at a

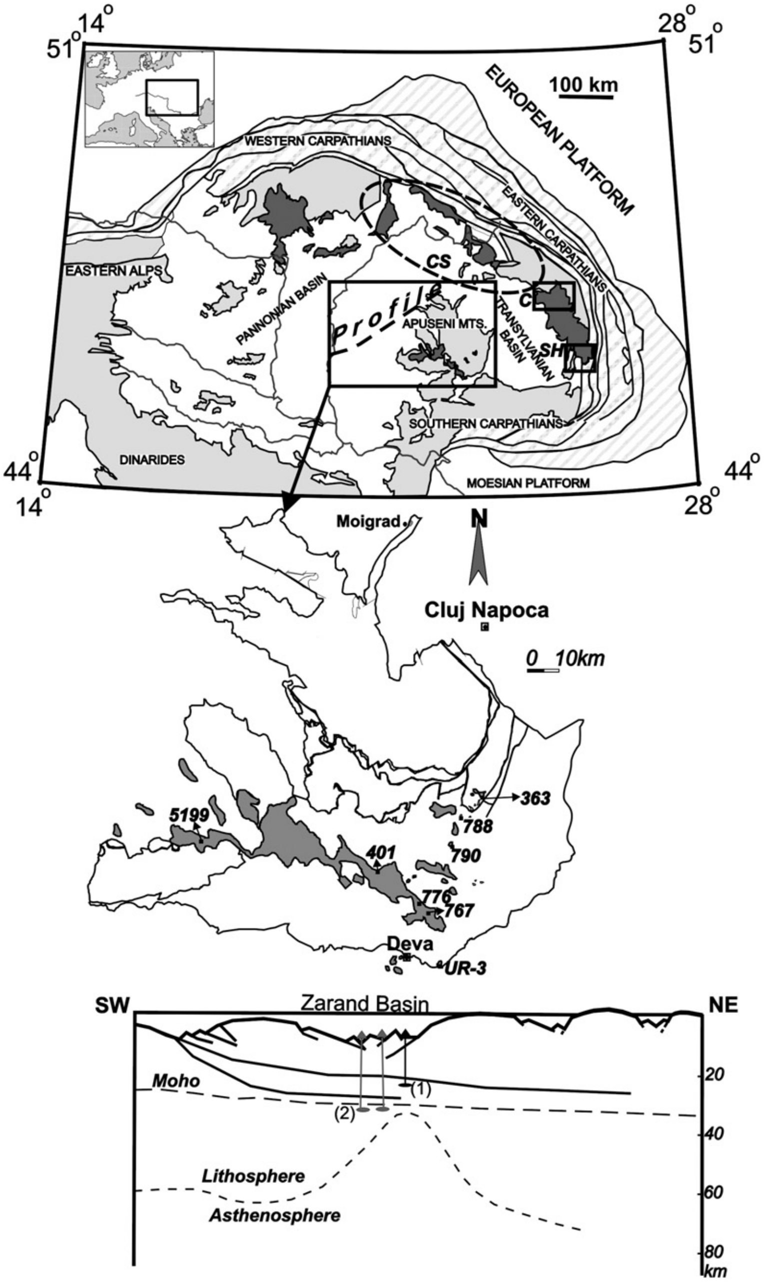

Fig. 1. Simplified sketch of Apuseni Mountains of western Romania showing the Neogene magmatic area (dark grey) and sample collection points. Inset shows the geographical setting, location of Central Segment (CS), Călimani (C) and South Harghita (SH) volcanic areas and location of the PGP-4 profile. Integrated interpretation of the low-frequency seismic reflection PGP-4 profile (Takács et al., 1996) indicates a major asthenosphere rise and strong lithosphere thinning below the Bekes and Zarand basins. Tectonic interpretation of the profile in Hungary is after Csontos et al. (2002) and in Romania 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 and 7 Ma.

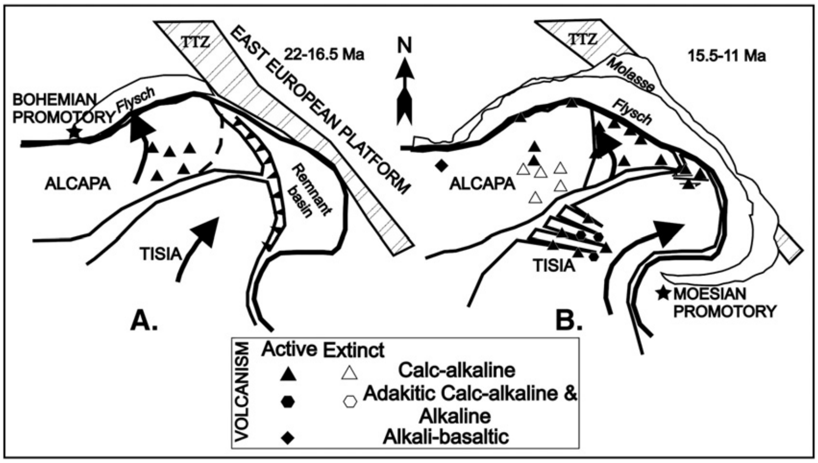

Fig. 2. Schematic examples of geodynamic evolution in Carpathian–Pannonian region during Middle to Late Miocene times from Seghedi et al. (2004) for two time-intervals 22–16.5 Ma and 15.5–11 Ma. Stars represent poles of rotation of the Alcapa and Tisia microplates. Clockwise rotation of Tisia around a pole situated in western Moesia (Csontos and Nagymarosy, 1998) facilitated thinning of the lithosphere and *en-echelon* brittle crustal splitting. Magmatism in the Apuseni Mountains is closely connected with the major eastward rotations and subsequent graben-type basin opening. Subduction of a remnant basin occurred towards the south-west and generated normal calc-alkaline volcanism along the Carpathian arc. Active and extinct volcanoes are simplified. TTZ = Tornquist–Teisseyre Zone.

distance of 200 km from the inferred subduction trench, which suggests that processes other than subduction must have been involved in magma generation (Fig. 2). 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 fluid involvement, the hydration state of the magmatic source, and the extent of crustal contamination 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

Table 1
Location, selected whole rock compositions and Mg# of rocks from the Apuseni Mountains, analysed in this study

Sample	Locality	Latitude	Longitude	SiO_2 (%)	K_2O (%)	Mg#	Ni (ppm)	Sr (ppm)	Y (ppm)	Nb (ppm)	Zr (ppm)	Age (Ma)	$^{87}Sr/^{86}Sr$ (wr)	$^{143}Nd/^{144}Nd$ (wr)
790	Citera	46.233	23.213	61.43	1.39	0.445	3.9	212	22.8	7.8	101	14.6±1.6	0.708312	0.512421
5199	Chisindia	46.278	22.081	60.56	1.30	0.469	14.5	249	28.5	6.3	118	12.8±0.6	0.704640	0.512778
767	Cetras	46.033	22.973	61.41	1.47	0.583	22	1005	19.6	9	101	11.7±0.5	0.704502	0.512634
401	Brad	46.138	22.844	63.58	1.89	0.397	5.8	321	29.3	8.1	106	11.0±0.4	0.705608	0.512593
776	Zambrita	46.045	22.953	58.09	2.92	0.623	56	2770	22.2	16.3	171	10.5±0.4	0.704250	0.512609
363	Poienita	46.326	23.286	61.70	1.99	0.508	7.3	665	21.2	17.4	156	9.3±0.4	0.704704	0.512667
788	Detunata	46.278	23.199	55.20	1.45	0.649	26.1	596	19.1	16.8	108	7.4±0.3	0.704400	0.512689
UR-3	Uroi	45.857	23.044	61.18	5.32	0.592	41	2353	16.5	24.9	317	1.6±0.1	0.704441	0.512680

Age determinations (K/Ar method) are from Pécskay et al. (1995) and Roşu et al. (1997, 2004b) and whole rock geochemistry and isotope compositions are from Roşu et al. (2004a).

lava compositions and the whole time interval of magma generation in the Apuseni province, for which whole-rock $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotope data were already available (Table 1).

Oxygen and hydrogen isotope ratios, in combination with $^{87}Sr/^{86}Sr$, provide powerful constraints on magmagenesis, particularly the extent of crustal contamination, as well as the possible effects of fluid involvement (e.g., distinguishing between melting due to fluid-related enrichment or slab–melt enrichment). 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, 2001) have shown a significant difference between the $\delta^{18}O$ 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 in between crystallisation of the phenocrysts and formation of the groundmass.

2. 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; Brezsnyansky et al., 2000; Neugebauer et al., 2001). The Tisia unit is identical to the “Tisza” unit discussed by Hungarian authors, e.g. Csontos et al. (1992, 2002). Tisia and Dacia are microplates with different Mesozoic tectonic histories (Surmont et al., 1990; Pătraşcu 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; Huismans 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 ~20° clockwise rotation (Pătraşcu 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°–50°(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 (~60°) (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 an NW–SE strike occurred along the western border of the Apuseni Mountains (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 (Ghiţulescu and Socolescu, 1941; Roşu et al., 1997; Balintoni and Vlad, 1998; Seghedi et al., 1998; Roşu et al., 2001, 2004a; Seghedi et al., 2004). A consequence of the clockwise rotation of Tisia was synchronous extensional (Györfi and Csontos, 1994; Tari et al., 1999) and compressional deformation (Huismans et al., 1997; Ciulavu et al., 2000), respectively west and east of the Apuseni Mountains. 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 (Ratschbacher et al., 1993; Bojar et al., 1998; Ciulavu et al., 2000; Huismans et al., 2001; Sanders et al., 2002).

Fig. 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) indicating an asthenospheric upwelling below the Zarand basin, the largest graben system at the western edge of the Apuseni Mountains. Fig. 2 shows interpretations by 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 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; Mațenco, 1997; Zweigel, 1997).

During Neogene times, contemporaneous with the Apuseni Mountains magmatism, much more extensive calc-alkaline magmatism also developed along the Carpathian arc, i.e. in front of the Alcapa and Tisia–Dacia microplates (Pécskay et al., 1995, 2006). This magmatism is thought to be related to subduction and probable breakoff of the subducted slab (Nemčok et al., 1998; Seghedi et al., 1998, 2001; Wortel and Spackman, 2000; Seghedi et al., 2004) and is situated much closer to the postulated Carpathian subduction trench (Fig. 2).

3. 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_2. Several temporal trends can be seen in the major element, trace element and isotopic compositions. Total alkalis, P_2O_5, MgO and $^{144}Nd/^{143}Nd$ ratios increase with time, whereas $^{87}Sr/^{86}Sr$ ratios decrease. Additionally, Nb/Y and Sr/Y ratios show a general increase from the onset to the end of magmatic activity (Fig. 3A, B). These trends suggest a tendency towards more alkaline magmatism and more "primitive" magma sources. Fig. 3C shows that all of the samples have typical calc-alkaline trace element characteristics (Nb- and Ti-depletion, strong enrichment in Pb). Two samples (alkaline trachyandesites 776 and UR-3) have conspicuously higher K_2O, Ba, Th and LREE contents. 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 youngest alkaline volcano (1.6 Ma) shows a trend towards an intraplate OIB-type composition (Fig. 3A,B).

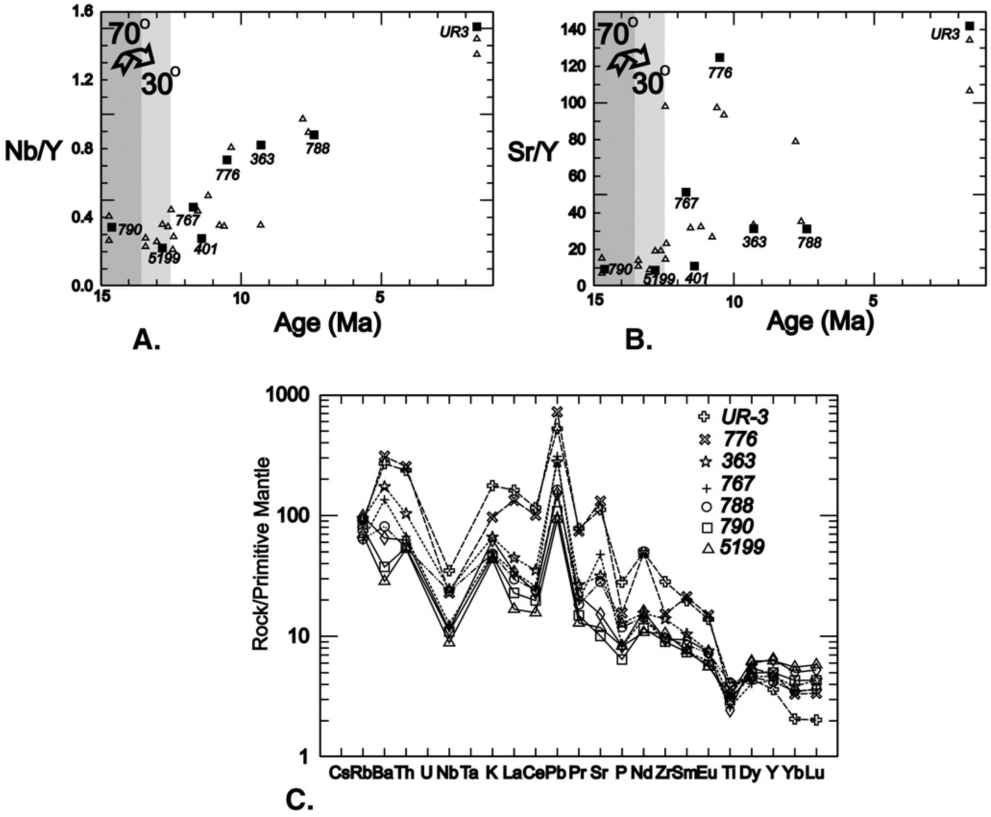

Fig. 3. (A) Nb/Y and (B) Sr/Y vs. age for Neogene magmatic rocks from the Apuseni Mountains (triangles), highlighting the samples used in this study (squares). Shaded areas represent 70° and 30° angle of declination suggesting main Miocene clockwise rotations in the Apuseni Mountains area (Panaiotu, 1998, 1999; Roşu et al., 2004a). (C) Primitive mantle normalized incompatible trace element diagrams for the studied rocks, using the normalizing coefficient of Sun and McDonough (1989); Data are from Roşu et al. (2004a).

Since among the normal calc-alkaline rocks, the older ones (14.5–13.0 Ma) have higher $^{87}Sr/^{86}Sr$ 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 $^{87}Sr/^{86}Sr$ ratio. In contrast, fractionation processes were less significant for the younger magmatic products: they show a narrow SiO_2 interval, no Eu anomalies, and have lower $^{87}Sr/^{86}Sr$ and higher $^{143}Nd/^{144}Nd$ ratios than the older ones. For rocks younger than 12 Ma, melting due to fluid-related enrichment 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).

4. Sample selection and analytical techniques

Geographic locations and key geochemical characteristics of samples examined in this study are given in Table 1. Petrographic data are given in the Appendix, 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 μm. Counting times were 30 s 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 20 W CO_2 laser following the technique of Sharp (1990). Oxygen was extracted from silicate minerals by fluorination with BrF_5 and was measured directly on a Finnigan MAT Delta Plus mass spectrometer without combustion to CO_2. Throughout the measurements several standards were analysed together with the samples. For standards and samples 1–2 mg of material were analysed. 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

Table 2
Chemical composition of cores (c) and mantles (m) of clinopyroxenes from selected Apuseni Mountains Neogene magmatic 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_2	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_2	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_2O_3	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_2O_3	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_2O	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

Earth and Planetary Sciences, University of New Mexico, Albuquerque, following the technique of Sharp et al. (2001). During the δD analysis, the international standard NBS 30 and an in-house standard Banco Bonido were analysed, for which values of −66±3‰ and −76±3‰ were measured, respectively. The water content was calculated from the intensity of the mass 2 signal. All the δ^{18}O and δD 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 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 0.14 ng of Sr. Measurements were made using a Finnigan MAT 262 multicollector thermal ionization mass spectrometer at the University of Pisa, Italy. Measured ^{87}Sr/^{86}Sr ratios were normalised to ^{86}Sr/^{88}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}Sr/^{86}Sr=0.710242±0.000013 (2SD). Results are given in Table 3.

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

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

Abbreviations: Amph—Amphibole; Bi—Biotite; Cpx—Clinopyroxene; Pl—Plagioclase; Gm—Groundmass.

5. Results

All the analysed 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. Reverse zoning in cpx could also be produced by interaction of the ascending magmas with peridotite (e.g. Gao et al., 2004). 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 δ^{18}O values of the mineral separates (Table 3) are plotted against SiO_2 content of the whole-rocks (Table 1). Clinopyroxene phenocrysts in all the measured rocks show an extremely restricted range of δ^{18}O values between 5.5 and 5.6‰. These values are slightly higher than the δ^{18}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 δ^{18}O values of clinopyroxene in mantle peridotite xenoliths worldwide (5.7‰; Mattey et al., 1994). There is no correlation between whole-rock SiO_2 content and pyroxene δ^{18}O values and also no variation of the clinopyroxene δ^{18}O values with age of the host rock. Some of the amphibole and biotite separates show similar or slightly higher δ^{18}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 δ^{18}O values for plagioclase and groundmass. Plagioclase phenocrysts show a wide variation of δ^{18}O values between 6.3 and 7.6‰. Even higher δ^{18}O values are given by the separated groundmass (6.7–10.1‰). The values for the groundmass are

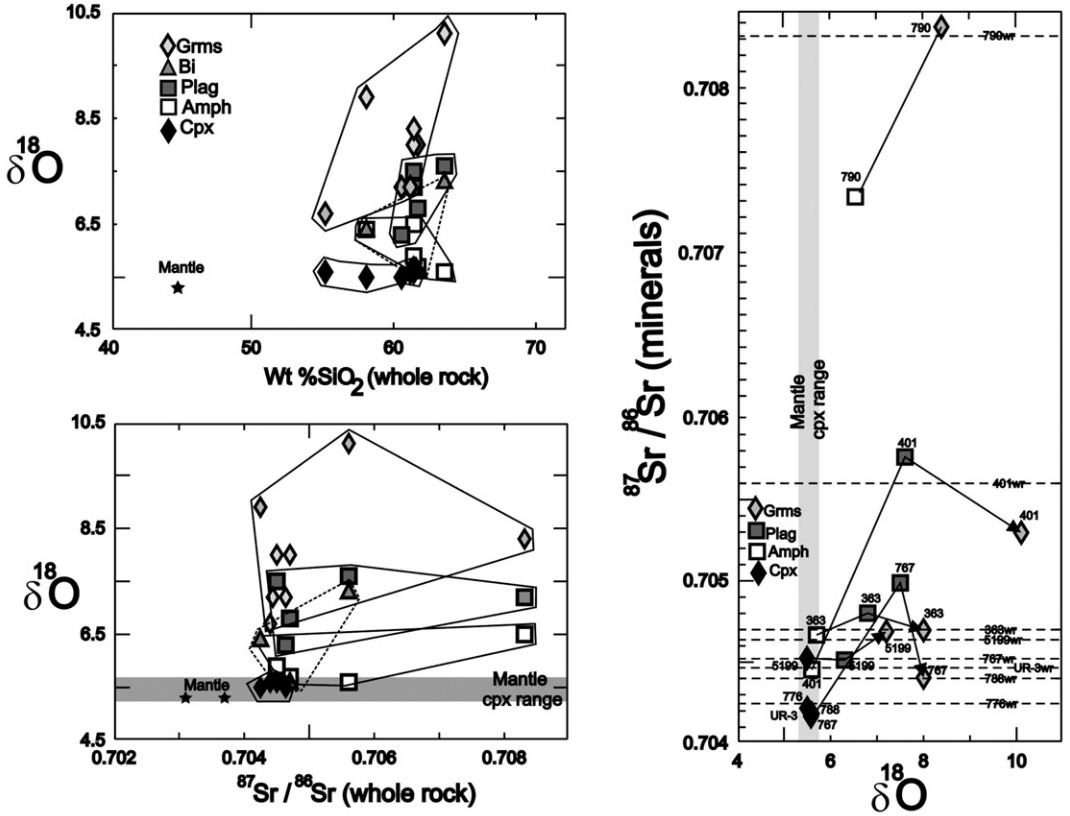

Fig. 4. A. $\delta^{18}O$ value of minerals and groundmass vs. whole-rock SiO_2; b. $\delta^{18}O$ value of minerals vs. whole-rock $^{87}Sr/^{86}Sr$; c. mineral $^{87}Sr/^{86}Sr$ vs. mineral $\delta^{18}O$ compositions for mineral separates and groundmass in selected Neogene magmatic rocks from the Apuseni Mountains. Stars represent the SiO_2 mantle composition and the whole-rock $^{87}Sr/^{86}Sr$ ratios for depleted and enriched mantle in Carpathian–Pannonian region, calculated from the composition of alkali basalts, as compiled by Mason et al. (1996).

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). Fig. 4B shows $\delta^{18}O$ mineral compositions plotted against $^{87}Sr/^{86}Sr$ ratios of the whole-rock. Most of the samples with low $\delta^{18}O$ values in their clinopyroxenes also have low whole-rock $^{87}Sr/^{86}Sr$ ratios, but two samples that do not contain clinopyroxene (790 and 401) have elevated $^{87}Sr/^{86}Sr_{WR}$ ratios (0.708 and 0.7056, respectively). Sample 790, representing the oldest magmatism at 14.6 Ma, contains amphibole with an elevated $\delta^{18}O$ 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 $^{87}Sr/^{86}Sr$ 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 $^{87}Sr/^{86}Sr$ and $\delta^{18}O$, but plagioclase and groundmass show a shift toward both higher $^{87}Sr/^{86}Sr$ and higher $\delta^{18}O$ values. The plot of mineral $^{87}Sr/^{86}Sr$ vs. mineral $\delta^{18}O$ values (Fig. 4C) shows consistently lower $^{87}Sr/^{86}Sr$ 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 $^{87}Sr/^{86}Sr$ ratios for mafic minerals from samples 363 and 5199 are close to the whole-rock $^{87}Sr/^{86}Sr$ ratio. The $^{87}Sr/^{86}Sr$ 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 $^{87}Sr/^{86}Sr$ ratio should be the weighted mean of the values for the separated minerals and groundmass, and Fig. 4C demonstrates that mafic minerals yield a 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 δD values on groundmass material vary from -84‰ to -127‰ (Table 3). They show a good correlation with the SiO_2 and age of the rocks, the older rocks having higher δD values (Fig. 5A and B). The amphiboles and biotites show a similar pattern. Generally, within a single sample, the groundmass has the lowest δD 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 Demény et al. (2006) who suggested that both high and low δD were produced by degassing (H_2O or H_2 release).

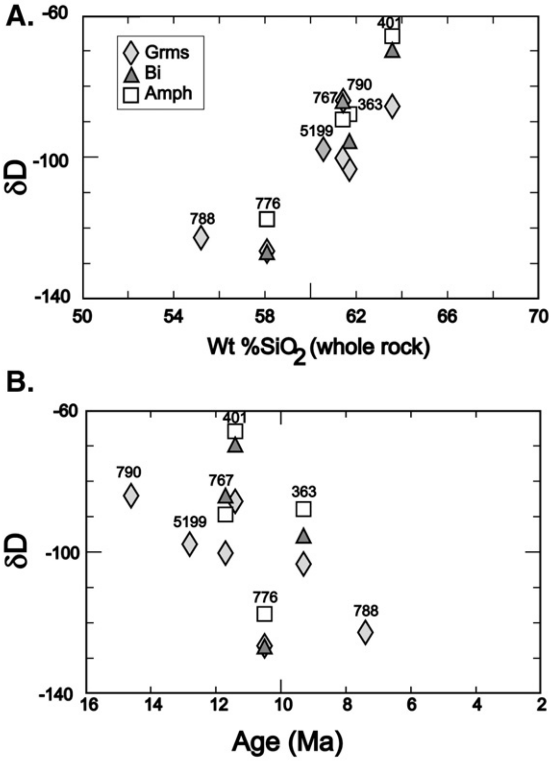

Fig. 5. A. δD of mineral phases and groundmasses vs. whole-rock SiO_2 in selected Neogene magmatic rocks from the Apuseni Mountains.; B. δD of mineral phases and groundmasses versus age of sample.

6. 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 and 10‰, the isotopic values of coexisting phases showing the following order of isotopic enrichment due to fractional crystallisation: biotite–hornblende–feldspar–groundmass (Taylor and Shepard, 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 $^{87}Sr/^{86}Sr$ between amphibole and plagioclase and also a quite large $\Delta^{18}O_{\text{amphibole–plagioclase}}$ of 2‰. This could indicate assimilation of material with higher $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ during and after crystallisation of plagioclase. All the other samples discussed in this paragraph fall in the group with generally low $^{87}Sr/^{86}Sr$ ratios; differences between them mainly concern the evolution of $\delta^{18}O$ values of the different phases. For sample 767, there is a large difference in $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ ratios between pyroxene and plagioclase, although the groundmass falls in the group with low values for $^{87}Sr/^{86}Sr$. 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 $^{87}Sr/^{86}Sr_{plag}$ as compared to the groundmass points towards assimilation during plagioclase crystallization. Tepley et al. (2000) suggested that the higher $^{87}Sr/^{86}Sr_{plag}$ compared with groundmass in Mexican volcanic rocks were due to recharge by a higher temperature magma with a lower $^{87}Sr/^{86}Sr$ ratio and higher Sr concentration.

Samples 776 and 363 show similar $^{87}Sr/^{86}Sr$ values between phases, although when considering the groundmass, they show a difference of >2‰ in $\delta^{18}O$. Samples 5199, 788 and UR-3 show similar values for $^{87}Sr/^{86}Sr$ for all the phases and $\delta^{18}O$ 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 δD 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 δD values characterizing the older samples and lower δD values for the younger ones (Fig. 5B). Accordingly, we can interpret the variation of δD values between samples as due to variation in δD of the source, with a more depleted source for the younger rocks. As well, δD data against the whole-rock SiO_2 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 $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ ratios similar to those of clinopyroxene; (2) a later melt phase with increasingly higher $\delta^{18}O$ and $^{87}Sr/^{86}Sr$, as suggested by the late crystallising phases (especially plagioclase and groundmass), indicating an effect of crustal assimilation. The restricted range of δD 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 $\delta^{18}O$ minerals have been removed and the residual liquid has also had its $\delta^{18}O$ 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 $\delta^{18}O$ 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 metalogenetic activity (Udubaşa et al., 2001, Roşu et al., 2004a,b, Neubauer et al., 2005), but the rocks in this study were unaffected by hydrothermal fluids.

7. Petrogenetic processes — Comparison with neighbouring areas

In is section, the Apuseni Mountains data will be compared with $\delta^{18}O$ 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. Fig. 6 shows the variations of $\delta^{18}O$ of pyroxene, amphibole and biotite versus whole-rock SiO_2 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

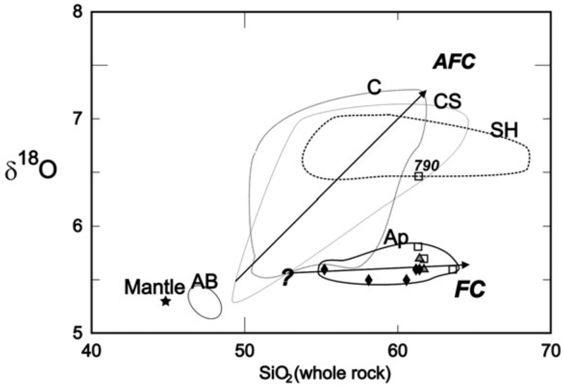

Fig. 6. $\delta^{18}O$ value of minerals vs. SiO_2 whole-rock values of several Neogene volcanic regions in the Carpathian–Pannonian region. Abbreviations: C—Călimani area; CS—Central segment volcanic area (as defined by Seghedi et al., 2004); Ap—Apuseni; SH—South Harghita volcanic area; AB—local alkali-basalts; 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).

volcanic fields in eastern Hungary, eastern Slovakia, southeast Ukraine and northern Romania (Fig. 1). For most of the Apuseni Mountains samples, the low degree of δ^{18}O variation over the range of 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 δ^{18}O values with increasing whole-rock 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 δ^{18}O values over a wider range of SiO_2 contents, however at higher overall δ^{18}O values.

The plot of the ^{87}Sr/^{86}Sr ratios for the mafic minerals against δ^{18}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}Sr/^{86}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 δ^{18}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 Fig. 7.

Low δ^{18}O and ^{87}Sr/^{86}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_2O contents) fall in the same field as primitive oceanic arc lavas (e.g. Eiler et al., 2000), revealing that the range of δ^{18}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.

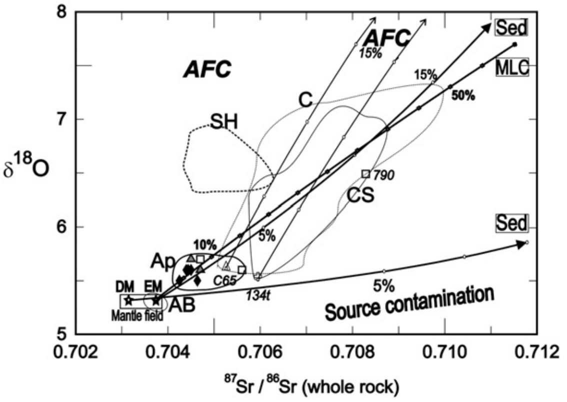

Fig. 7. δ^{18}O value of minerals vs. ^{87}Sr/^{86}Sr ratios for several Neogene volcanic regions in the Carpathian–Pannonian region (^{87}Sr/^{86}Sr values for Apuseni Mts are mafic mineral values, whereas those for the other regions are whole-rock values). Abbreviations: C—Călimani area; CS—Central 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% 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 134 t) and an upper 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.

The Apuseni and South Harghita magmas show a similar restricted range of low $^{87}Sr/^{86}Sr$ ratios (0.7045–0.706) that may suggest a depleted source for both areas, but with much lower $\delta^{18}O$ values for the Apuseni Mountains. The shift to higher $\delta^{18}O$ values in South Harghita, combined with the small variation in $^{87}Sr/^{86}Sr$ 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 $\delta^{18}O$ values (6–6.9‰) and melts from a variably enriched mantle source.

8. 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 granulite xenoliths have been brought to the surface by alkali basaltic magmas in the neighbouring Pannonian Basin (Kempton et al., 1997; Dobosi et al., 2003; Embey-Isztin et al., 2003). Their isotopic characteristics are extremely unusual for continental lower crust. Some LREE-depleted granulites have MORB-like ε_{Nd} values and $\delta^{18}O$ values as low as 3.8–5.2‰. They have been interpreted as representing accreted altered oceanic crust possibly of Mesozoic age (Dobosi et al., 2003). Although they represent the lower crust of the Alcapa terrane rather than the Tisia–Dacia block, it is possible that such material has also been accreted beneath the Apuseni Mountains, particularly as Mesozoic ophiolites are well known in the region (Bortolotti et al., 2002). Along with mafic granulite xenoliths, metasedimentary lower crustal material has been found, whose isotopic composition was already used 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 $\delta^{18}O$ 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 $\delta^{18}O$ variation for volcanic rocks from the Călimani and Central Segment volcanic areas; b) large Sr/Y variation with a narrow range of $\delta^{18}O$ 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 $\delta^{18}O$ variation, correlating with low $^{86}Sr/^{87}Sr$ ratios (c.a. 0.7045), with higher K_2O 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 $\delta^{18}O$ values to those from the Apuseni samples (Bindeman et al., 2005). On the other hand a narrow range and low $\delta^{18}O$ 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 $\delta^{18}O$, 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 δD values, suggesting a change in source composition (Fig. 5).

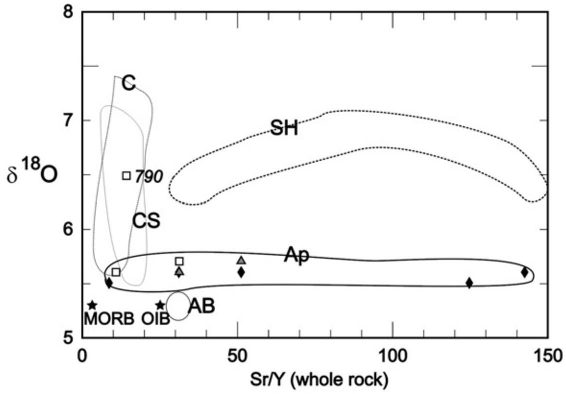

Fig. 8. $\delta^{18}O$ values of minerals vs. Sr/Y whole-rock compositions of several Neogene volcanic regions in the Carpathian–Pannonian region. Abbreviations: C—Călimani area; CS—Central segment volcanic area (defined by Seghedi et al., 2004); Ap—Apuseni; SH—South Harghita volcanic area; AB—local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

We have used a plot of $\delta^{18}O$ mineral compositions vs. Nb/Zr in the whole-rocks (Fig. 9), since this ratio

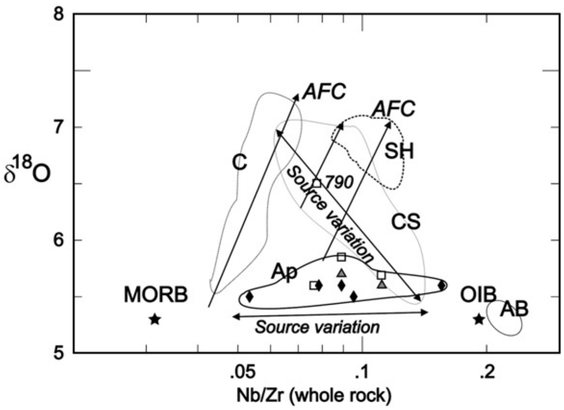

Fig. 9. $\delta^{18}O$ values of minerals vs. Nb/Zr whole-rock compositions of several Neogene volcanic regions in the Carpathian–Pannonian region. Abbreviations: C—Călimani area; CS—Central segment volcanic area (defined by Seghedi et al., 2004); Ap—Apuseni; SH—South Harghita volcanic area; AB—local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

gives constraints on different mantle source components varying from ~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 $\delta^{18}O$ 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 large variation in Nb/Zr ratios and very little variation in $\delta^{18}O$ compared to subduction-related magmas from elsewhere in the Carpathian–Pannonian region. 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 $\delta^{18}O$ 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).

9. 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; Martin et al., 2005); 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).

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 (Fig. 2) 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}Sr/^{87}Sr ratios, the highest amphibole δ^{18}O and the lowest groundmass δD 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}Sr/^{86}Sr and δ^{18}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}Sr/^{86}Sr and δ^{18}O values than the older ones. This was probably following a period of increasing temperature at the base of the crust due to rotational shear heating at the termination of the period of fast rotation, which converted low δ^{18}O 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}Sr/^{86}Sr and δ^{18}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 an adakitic character (lower Sr/Y), as well as lower ^{87}Sr/^{86}Sr and δ^{18}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. A major asthenosphere rise accompanied by lithosphere thinning below the main volcanic area of the Apuseni Mountains is indicated by the low-frequency seismic reflection PGP-4 profile (Fig. 1).
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}Sr/^{86}Sr and δ^{18}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.

10. Summary and conclusions

Volcanic rocks from the Apuseni Mountains Neogene magmatic province have δ^{18}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}Sr/^{86}Sr ratios for clinopyroxenes (~0.7041) and amphiboles (0.7042–0.7042) are lower than whole-rock ^{87}Sr/^{86}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 δ^{18}O, δD and ^{87}Sr/^{86}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 δ^{18}O, δD and ^{87}Sr/^{86}Sr in their mafic phenocrysts, which correlate with the narrow range and low values of whole-rock ^{87}Sr/^{86}Sr ratios, different from most other calc-alkaline volcanic rocks in the Carpathian–Pannonian region. This suggests a source that had a rather uniform δ^{18}O composition with chemical heterogeneities, such as a lower crust composition. This enables us to reject the hypothesis of slab–melt processes (invoked for South Harghita) and of a mantle-source influenced by subduction-related metasomatism, as has been suggested for all the other Neogene–Quaternary magmas in the Carpathian Pannonian area (except South Harghita) that are mostly chemically and isotopically homogeneous (e.g. Seghedi et al., 2004). 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 δ^{18}O and ^{87}Sr/^{86}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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Appendix A. Petrography

The effusive 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. Andesites and porphyritic microdiorites occur as clusters of small intrusive bodies associated with the effusive rocks. The spatial distribution of different types of andesites seems to follow a systematic trend with more basic pyroxene-bearing varieties in the west, and more acidic, amphibole-and biotite-bearing varieties in the east. Intermediate-type, amphibole–pyroxene andesites occur in between.

Basaltic andesites are present as two small occurrences in the Detunata hills (*sample 788*) and also occur in the Zarand area. They are slightly porphyritic, with plagioclase (labradorite to anorthite), clinopyroxene and rare amphibole microphenocrysts in a groundmass of plagioclase microlites, clinopyroxene, magnetite, ilmenite and glass.

Andesites display a large spectrum of varieties. Two-pyroxene andesites prevail in the Zarand Mts. (*sample 5199*) (Fig. 1). They are porphyritic and contain phenocrysts of plagioclase (andesine to bytownite), clinopyroxene and orthopyroxene. The groundmass consists of plagioclase and clinopyroxene microlites, magnetite, ilmenite and glass. Amphibole ± pyroxene andesites (*sample 790*) are normally to strongly porphyritic and have plagioclase (andesine to bytownite), amphibole, rare clinopyroxene, and sporadic corroded quartz and accessory Fe–Ti oxides, apatite, zircon and Cr-spinels. Amphibole is partially opacitised. Rare garnet-bearing varieties can be found at Zlatna, Bucium and in the Brad areas, showing secondary hydrothermal alteration (propylitic), as secondary chlorite on amphibole or pyroxene. Amphibole–biotite ± pyroxene andesites (*samples 363, 401 and 767*) are strongly porphyritic. Main phenocrysts are plagioclase (andesine to anorthite), amphibole, biotite, clinopyroxene and as accessory minerals Fe–Ti oxides, apatite, zircon and Cr-spinels. These rock-types often contain quartz phenocrysts. Amphibole and biotite are partially opacitised.

Dacite are found mostly in the Roşia Montană area, showing porphyritic textures and hydrothermally altered plagioclase, quartz, amphibole and biotite as phenocrysts, and magnetite, ilmenite, apatite and zircon as accessory minerals in a microgranular or glassy groundmass.

Besides the above-mentioned lithologies, characterized by normal calc-alkaline compositions, there are a few small-scale occurrences of alkaline trachyandesite rocks in the south-eastern part of the region. One occurs at Zâmbriţa (*sample 776*) and the second at Uroi (*sample UR-3*) in the south-eastern extremity of the region (Fig. 1). Zâmbriţa trachyandesite is slightly porphyritic and contains plagioclase (labradorite to anorthite), amphibole, clinopyroxenes, biotite, and quartz as microphenocrysts in a microgranular groundmass of the same composition, with accessory apatite, zircon, magnetite, ilmenite and Cr-spinel. The Uroi trachyandesites (according to TAS diagram) or shoshonites (according to K_2O vs. SiO_2 diagram) closely resemble the shoshonites that occur at the southeastern end of the East Carpathian volcanic range (e.g. Seghedi et al., 2004). The rocks are slightly porphyritic and display a mineral assemblage containing plagioclase (andesine to anorthite), clinopyroxene > orthopyroxene, amphibole and biotite microphenocrysts in a microgranular groundmass of the same composition that contain apatite, zircon, magnetite and ilmenite as accessories. Amphibole and biotite are partially opacitised and resorbed. Accidental quartz xenocrysts or clots of plagioclase, amphibole and biotite are present.

A petrographic description for each studied sample is given in the following table:

Sample	Locality	Color	Structure	Texture	Mineral composition Main minerals	Mineral composition Accessory minerals	Groundmass	Comments
790	Citera	Greyish-black	Massive	Porphyritic	Pl (45–50%), 5/1 mm; Amph (10–15%), 8–10/4–6 mm; Cpx (3–4%) 2/0.9 mm; Opx (1–2%) 0.5/0.3 mm	Apatite, zircon, magnetite (3–5%)	Glassy to microgranular (30–40%)	Amphibole is partially opacitised
5199	Chişindia	Greyish	Massive	Porphyritic	Pl (40–45%), 3/1 mm; Cpx (14–16%) 3/0.9 mm; Opx (4–6%) 1.5/0.5 mm	Apatite, (zircon), magnetite, (ilmenite) (2–5%)	Glassy (25–35%)	
767	Cetraş	Pale grey	Massive	Strongly porphyritic	Pl (50–55%), 7–15/3–5 mm; Amph (20–25%), 4–7/2–3 mm; Cpx (3–4%) 3/1 mm; Bi (4–6%); Quartz (2–3%), 4–12/2–5 mm	Apatite, zircon, rutile, magnetite, chrome–spinel (4–6%)	Glassy to microgranular (10–20%)	Amphibole and biotite are partially opacitised. Quartz is partly corroded and show a violet color
401	Brad	Pale grey	Massive	Porphyritic–strongly porphyritic	Pl (40–45%), 5–10/4 mm; Amph (5–7%), 4–5/1.5–2 mm; Bi (2–4%), 7–10 mm; Quartz (3–4%), 4–6/2–4 mm	Apatite, zircon, magnetite, (ilmenite) (2–4%)	Glassy (30–45%)	Amphibole and biotite are partially opacitised. Quartz is corroded
776	Zâmbrița	Dark grey or black	Fluidal or massive	Slightly porphyritic	Cpx (5–7%), 0.5–2 mm; Amph (5–8%), 0.5–1/0.1–0.2 mm; Bi (3–5%), 4–6 mm; Pl (65–70%), 0.5–0.7 mm most as microlites	Apatite, zircon, rutile; magnetite (ilmenite), chrome–spinel (5–7%)	Glassy to microgranular (40–60%)	Amphibole and biotite are partially opacitised.
363	Poienița	Pale grey	Massive	Strongly porphyritic	Pl (45–50%), 7–15/3–5 mm; Amph (25–30%), 6–11/3–5 mm; Cpx (0.5–1%) 2/0.5 mm; Bi (4–6%), 3–5 mm; Quartz (1–2%), 4–6/2–3 mm	Apatite, zircon, magnetite, ilmenite (3–5%)	Glassy to microgranular (10–20%)	Amphibole is partially resorbed
788	Detunata	Dark grey or black	Massive or fluidal	Aphyric	Cpx (25–30%), 0.5–2 mm; Amph (<1%); Pl (55–60%), 0.2–0.7 mm as microlites;	Magnetite, ilmenite (5–6%)	Glassy to microgranular (70–80%)	Amphibole is resorbed
UR-3	Uroi	Grey or reddish	Massive or fluidal (trachytic)	Aphyric	Cpx (8–10%), 0.3–1.2 mm; Amph (3–4%), 0.5–1/0.1–0.2 mm; Bi (2–4%), 4–6 mm; K-feldspar (50–65%), 0.1–0.3 mm as microlites	Apatite, magnetite, (pseudo–brookite), ilmenite (2–3%)	Glassy to microgranular (70–80%)	Amphibole and biotite are opacitised and resorbed.

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