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Petrogenesis and magmatic evolution in the East Carpathian Neogene volcanic arc (Romania)

I. Seghedi[1], A. Szakács[1], P. R. D. Mason[2,*]

1. *Geological Institute of Romania, str. Caransebeş 1, RO-78344 Bucharest 32, Romania*
2. *Department of Geology, Birkbeck College, University of London Malet St., London, WC1E 7HX, UK*

Abstract

Along the inner part of the East Carpathians, a discontinuous subduction-related volcanic arc including the Oaş-Gutâi (OG), Ţibleş-Toroiaga-Rodna-Bârgău (TTRB), and Călimani-Gurghiu-Harghita (CGH) segments, developed during the Neogene-Quaternary. Based on extensive petrographic geochemical and isotopic data, the complex petrogenetic processes in this arc can be summarized as follows: (1) magma with a «subduction-related» chemical signature is the common source for all the segments. Two mantle sources were recognized: one for the South Harghita volcanics and the other one for the remainder of the arc segments. An additional crustal source is suggested for the northern segments (OG, TTRB and Călimani from CGH); (2) mantle-derived magmas were formed by almost the same degree of partial melting (10-15%), whilst South Harghita magmas result from gradually decreasing degrees of partial melting along the chain; (3) in areas with large volumes of erupted magmas (Călimani, Gutâi,) fractional crystallisation combined with crustal assimilation are the dominant petrogenetic processes. In areas with smaller volumes of erupted products (Oaş, Ţibleş, Toroiaga, Rodna) extensive mixing between crustal-derived magmas in deep-seated magma chambers is envisaged. The South Harghita volcanics suggest a complex combination of fractional crystallisation, magma mixing and assimilation, characteristic for each volcanic structure.

1. Introduction

The East Carpathian volcanic arc (ECVA) is an important geological structure in the East Carpathians. The volcanic activity accompanied the complex tectonic movements during Lower Miocene-Pleistocene times. The arc was formed by southward and westward subduction of the Eurasian Plate beneath small continental fragments, followed by continental collision of the Afro-Arabian and European Plates (Săndulescu, 1988; Royden & Burchfiel, 1989).

The ECVA volcanism, which is mainly calc-alkaline with minor shoshonitic series rocks, shows a wide range of petrographic types and geochemical and isotopic variations (e.g. Peltz et al., 1974; Peccerillo & Taylor, 1976a; Borcoş et al., 1979; Berza et al., 1982, 1984; Udubaşa et al., 1983; Seghedi et al., 1986, 1987; Kovács et al., 1992; Mason et al. , 1995; Mason et al., in press). We present a general picture of the origin and evolution of the ECVA magmas and focus on the implications of important petrogenetic processes. We use extensive published and unpublished petrographic, geochemical and isotopic data. The study does not include the Lower Miocene (Badenian) altered explosive acidic rocks, as the volcanic centers of these rocks have not yet been identified in Romania.

2. Distribution of Neogene – Quaternary magmatic rocks in eastern Carpathian volcanic arc

The ECVA lies along the inner part of the Romanian East Carpathians (Fig. 1). Large occurences of calc-alkaline volcanism are clustered from north to south in three principal segments: (1) Oaş-Gutâi (OG); (2) Ţibleş-Toroiaga-Rodna-Bârgău (TTRB); (3) Călimani-Gurghiu-Harghita (CGH).

The OG segment is controlled by NW-SE, N-S and W-E-oriented deep fracture zones. In Oaş, calc-alkaline rock assemblages are complex and from mainly isolated

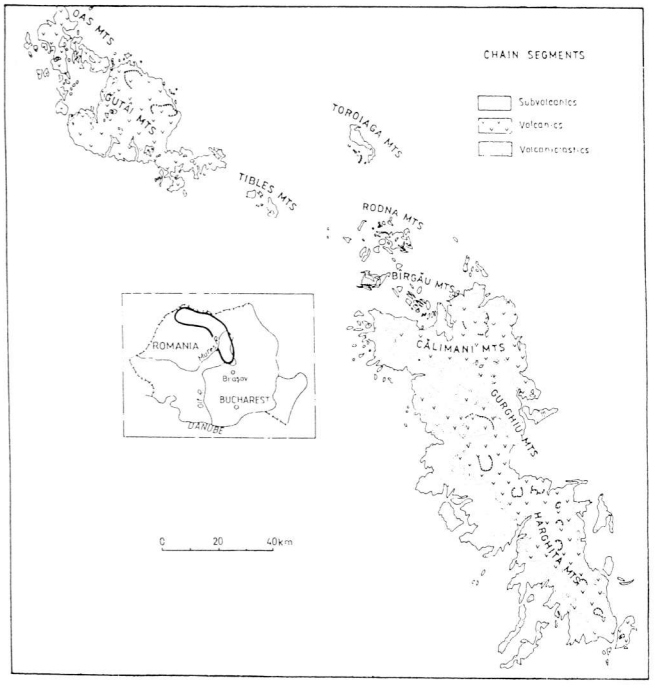

Fig. 1 – Sketch-map of the distribution of Neogene-Quaternary magmatic segments in the East Carpathians (Romania).

* Present address: NERC ICP-MS Facility, Centre for Analytical Research in the Environment, Imperial College, Silwood Park, Ascot, Berkshire, SL5 7TE, UK.

structures. Petrographic types range from basaltic andesites to rhyolites. K-Ar data (Pécskay et al., 1995) suggest that rhyolites and dacites pre-date the andesites and basaltic-andesites between 12 and 9 Ma. In the Gutâi Mts., volcanic edifices are superimposed upon one another and actually deeply eroded. Volcanic rocks are calc-alkaline basalts, basaltic-andesites, andesites and dacites, with a predominance of two pyroxene-bearing andesites (Kovács et al., 1992). Older volcanics are dacites in the south (13 Ma) (Pécskay et al., 1994) and younger basalts in the central area (7-8 Ma) (Edelstein et al., 1993).

The TTRB sector or «subvolcanic zone» is characterised by shallow intrusive bodies. Rock types range from calc-alkaline basalt to rhyolite. Rhyolite and dacites are generally older than basalts and basaltic andesites, the main interval of intrusions being 9 to 11 Ma (Pécskay et al., 1995).

The CGH arc sector consists of a 160 km long continuous volcanic range, with decreasing width, height and volume from north to south. Volcanic activity formed stratovolcanic edifices, building up a chain of adjoining and partially overlapping composite volcanoes surrounded by volcaniclastic aprons (Szakács & Seghedi, 1995). Typical calc-alkaline rocks of the CGH, range from basalts to rhyolites, but andesites form most of the chain (Rădulescu, 1973; Peltz et al., 1974). Some volcanics in the Călimani Mts. show a low-K calc-alkali trend (Seghedi, 1987) and rocks in the southernmost segment of the chain are shoshonitic in composition (Seghedi et al., 1986; 1987). Volcanic activity took place between 9.5-0.2 Ma ago. A spectacular age progression is seen for 40 km along the southernmost chain segment (4.5-0,2 Ma) (Szakács et al., 1993; Pécskay et al., 1995; Szakács & Seghedi, 1995).

3. Petrography

In the ECVA, lithologies range from basalts to rhyolites, although andesites are most prevalent. There are petrographic differences between different arc segments, but each contains most of the petrographic range (Tab. 1).

Effusive rhyolites are extensive only in the Oaș Mountains and are slightly porphyritic with plagioclase, biotite and quartz microphenocrysts in a mostly glassy matrix. The Călimani Dragoiasa volcanics are similar with a slightly rhyolitic character (Peccerillo & Taylor, 1976a; Nițoi, 1986; Mason et al., 1995), but are mainly dacites (Nițoi, 1986; Seghedi, 1987). Similar petrographic features, but with a microcrystalline matrix, are seen in rhyolites from the Rodna Mountains.

Dacites are the second most abundant rock type in the ECVA. They are classified on the basis of their mafic phenocryst assemblage: with orthopyroxene and glassy matrix (Oaș and Gutâi Mts.); with amphibole and pyroxene (Oaș, Gutâi, Rodna and North Harghita Mts.); with amphibole, biotite and quartz (Rodna, Toroiaga, Țibleș, Călimani, South Harghita Mts.); with biotite, amphibole, quartz, two pyroxene and olivine (Gutâi, Călimani, Harghita Mts.). This latter variety suggests magma mixing between acid and basic end-members. Garnet (almandine) is also present in some amphibole-biotite dacites from Rodna intrusives.

Andesites with two pyroxenes, two pyroxene and amphibole are the most dominant. A few aphyric andesites and dacites occur in several volcanic centers of the CGH segment. Basalts with clinopyroxene and olivine are present in the Gutâi, Călimani, Rodna and Bârgău Mountains. Important accessory minerals in the calc-alkaline series include magnetite, ilmenite, apatite and zircon (the latter mainly in acidic rocks).

Small volume shoshonitic rocks in the southernmost part of the CGH contain plagioclase, K-feldspar, two pyroxenes, amphibole, biotite, olivine and quartz, in a non-equilibrium mineral assemblage. This is characteristic of magma mixing between basic and acidic end-members (Seghedi et al., 1987; Mason et al., in press).

Crustal xenoliths (metamorphic and sedimentary rocks from the basement) are generally rare, but cognate xenoliths, which represent accumulation of crystal clots (plagioclase + amphibole + pyroxene + magnetite), are widespread in all the areas.

Table 1 – ECVA groups of selected samples for which major, trace element data and Sr isotope analyses are presented in Table 2. For each group sample number, petrography and location are shown. Analyses marked * were performed in the U.K. Abbreviations: B-basalts; BA-basaltic andesites; A-andesites; D-dacites; RD-rhyodacites; R-rhyolites; Sh-shoshonites; Di-diorites; Gd-granodiorites; Mzd-monzodiorites; Mzg-monzogabbros; G-gabbros; af-aphanitic; h-hyalo-; m-micro; px-pyroxene; am-amphibole; ol-olivine; bi-biotite; q-quartz.

OAȘ Mountains			GUTÂI Mountains		
Sample no.	Petrography	Location	Sample no.	Petrography	Location
BO-75	R	Orasul Nou	BM-105	R bi	Danesti
BO-72	R	Orasul Nou	BM-95	D am	Plesca Mare
208	R px,am	Remetea	BM-106	D am,bi	South Danesti
225	A px	Piatra Socii	BM-25	A px	Higea Ridge
2002	mDi px	Calinesti	BM-36	A px	Ciurca Noua Valley
222	A am,px	Barlog	BM-39	A px	Baia Sprie
210	A px	Jeleznic	BM-32	A px	Suior
216	A am,px	Custurii Valley			
BO-88	A px	Tarnei Valley			
TIBLEȘ Mountains			TOROIAGA Mountain		
BT-37	RD	Hudin	14	A q,bi,am	Baia Borsa Quarry
BT-30	mGd	Paltinis Peak	13	A am,px	Fataciuni Creek
BT-40	D	Buza Mlastinilor Peak	185	A am,bi	Colbu Creek
BT-15	Mzd	North Tomnatec Peak	86	Di q,px,am,bi	Tiganul Creek
BT-12	Mzg	North-East Tibles Peak	518	BA px,am	Arsita Creek
BT-8	Mzd	Bran Peak			
BT-24	G	Negru Stream			
BT-7	mG	South Arsuri Peak			
BT-4	G	Magura Neagra			
RODNA Mountains			BÂRGĂU Mountains		
397	R bi	Borta Peak	C46*	BA af	Dorna Quarry
549	D am	Magura Sturzii	C44*	BA am,px	Dorna Borcut
273	D am,bi	Sangeorz Bai			
537	A am,bi	Vinului Valley			
538	A am,bi	Vinului Valley			
539	A am,px	Cozasel Valley			
224A	BA px,am	Izvorul Plesilor			
545	BA px	Magura Ilvei			
544	B px,am	Cormaita			
CĂLIMANI Mountains			GURGHIU Mountains		
1123	D bi,am	Bolovanis Valley	G15*	A am	Salard Valley
C13*	D am,bi,px,ol	Pietricelul Peak	330	A am,px	Fancel Ridge
C22*	A px,am	Tomnatec Valley	135	A am	Jirca Valley
5036	A px,am	Fantanelul Valley	G7	A px	Padurea Cetatii Ridge
1500	A px	Piatra Dornei	G39*	A px,am	Sobasa Creek
1492	BA px	Prislop Valley	G124A	A px,am,ol	Kemenes Koves
5158	BA ol	Tarnita Ridge	G32*	A px,am	Iuhodul Draculu Valley
3377	BA ol,px	Patului Creek	183	A px	Magura Ridge
1925	BA px	Jlisoara	G19	A px,am	Sumuleu Mare Valley
NORTH HARGHITA Mountains			348	B px,ol	Zespezel Peak
H28*	D af,am	Rachitis Peak	SOUTH HARGHITA Mountains		
H27*	D af	Var Valley	CP/1P	D am,bi	Olt Valley
86	A am,px,bi	East Harghita Ciceu	H47*	D bi,am	Bradului Valley
H33*	A px	Stanca Bufnitei	C82	A px,ol	Koves Valley
826	A px,am	Filio Ridge	837	A px	West Tekero Creek
H29*	A px,am	Liban Quarry	C77	A am,bi	Cucu Ridge
LV73	A px	Harghita Sicului	C76	A am,px,bi	Podul de Piatra
			H3*	Sh	Bicsad
			L18	A px	Harom Peak
			H11*	BA px	Mitaci Creek

4. Analytical techniques

Over 700 samples were collected to investigate the major lithologies within the ECVA. Selected major and trace element data and Sr isotope analyses are shown in Tab. 2.

Major elements were determined by wet chemical methods at Prospecțiuni S.A. laboratory. Trace elements have been determined by emission spectrography at the Geological Institute of Romania. The detection limits were 1 ppm for Yb, 3 ppm for Cu, Pb, Zn, Cr, Co, Ni, Sc, V and 10 ppm for Nb, Zr, Y, Sr, Ba. Rb and Sr

Table 2 – Major and trace element data and Sr isotope analyses of ECVA magmatic rocks. The determination made in U.K. were calculated on a volatile-free basis and all Fe was determined as Fe_2O_3. Major elements are given in wt% and trace elements in ppm.

	OAŞ Mountains									GUTÂI Mountains						
	BO-75	BO-72	208	225	2002	222	210	216	BO-88	BM-105	BM-95	BM-106	BM-25	BM-36	BM-39	BM-32
87Sr/86Sr	0.7071	0 7057	0 7083	0.7120	0.7089	0.7107	0 7043	0 7087	0.7066		0.7090		0 7083	0.7080		
SiO2	75.71	75.35	71.92	67.57	66.37	64.56	61.75	59.06	54 24	70 24	68.90	65 55	58.56	57.88	55.69	50.55
TiO2	0.16	0.20	0.42	0.38	0 44	0.62	0.86	0.80	1.00	0.50	0.40	0.72	0.70	0.76	0.76	0.98
Al2O3	13.40	12.98	15.02	15.93	15.48	15.79	15.24	16.89	19 46	14 40	14.86	15.34	17.61	18.00	16.53	18.75
Fe2O3	0.50	0.85	1.55	1.76	4.00	3.25	4 10	4.29	3 42	1 77	2.02	2.74	4 59	5.61	2.90	6.12
FeO	0.00	0.00	0.05	1.40	0.59	1.85	1.44	2.74	4.29	0.54	1.57	1.55	3.03	2.16	4.97	4 09
MnO	0.00	0.02	0.05	0.09	0 08	0.11	0.10	0.14	0 18	0.07	0.08	0.08	0.13	0.14	0.16	0.16
MgO	0.17	0.22	0.24	0.48	0.77	2.05	2 34	3.22	1.92	0.26	1 85	1.40	2.80	2.61	4.75	4 65
CaO	1 01	1.31	2.00	3 62	3.27	4 54	5.48	6.98	8.95	4.97	2.95	4.44	6.70	7.26	8.40	9.50
K2O	4.37	4 24	3.04	2.65	3.08	2.51	2.02	2.00	1 36	1.83	2.78	2.46	1.63	1.23	0.97	1.02
Na2O	2.91	3.10	3.71	3.82	3.73	3.16	3.21	2.68	2.73	3.00	2.60	3 52	2.62	2.92	2.33	2.26
P2O5	0.08	0.04	0.18	0.11	0 13	0.12	0.17	0.13	0.00	0.15	0.02	0.13	0.15	0 20	0.13	0.15
H2O+	1.06	1.10	1.52	1 84	1.61	1.09	1 47	1.17	2.03	1.46	0.70	1.55	1.02	0 94	2.00	1.32
S	0.28	0.16	0.08	0 07	0.08	0.07	0.07	0 08	0.18	0.08	0.03	0.12	0.17	0.16	0.29	0.14
Total	99.89	99.71	99.78	99.72	99 63	99.72	100.25	100.18	99.92	99.67	97.86	99.6	99.71	99.93	99.88	99.69
Sc	4	4	6	10	16	13	18	18	17	12	12	11	12	19	26	24
V	4	3	22	19	14	80	135	130	160	60	63	88	75	140	200	225
Cr	2	3.5	4			6	13	26	5.5	13	18	8	3	3	85	15
Co	2.5		3	3	2.5	6.5	8.5	9	11	4.5	6.5	8	8	13	22	17
Ni	3	7	5.5	7	3.5	6	11	11	9	17	7	6	7.5	5.5	20	14
Cu	5.5	17	10	21	5.5	13	13	21	13	8	9.5	4	5	11	60	30
Zn			42	75	70	78	94	78	63	50	65	50	73	125	90	95
Ga	15	17	15	18	14	13	16	17	12	20	15	15	12	15	20	11
Rb	170	200	100	100	110	100	70	90	50	51	140	98	60	40		35
Sr	110	130	160	220	210	195	245	260	375	280	280	240	235	210	185	225
Y	26	28	33.4	31 8	38 4	23 1	32 3	21.7	29 0	26	24	24	20	28	22	24
Zr	130	140	171	197	217	136	180	127	130	140	170	135	70	145	130	43
Nb	12	12	20.5	18.4	19.3	16.2	22.4	15.8		16		15				
Ba	700	830	610	670	600	500	550	500	370	450	730	460	300	260	240	180
La	32	35	40	35	38						36					
Pb	11	21	14	15	13	7.5	10	16	10	8 5	32	7	2	6.5	46	2
Yb	2.6	2.9	3 2	3.6	3.5	2.4	2.8	1 9	2 4	2.4	3	1.7	1 2	3.5	2.9	1.2
Th	14 8	12	9.5	11 3	14 3	9	6.2	10.7	5 5	5 6		7	7.1	4.2	4 6	1.3
U	2 6	3 2	3 3	3 7	2 9	3.7	2.1	2 3	1.7	1 8		2 8	1.8	0 9	1 3	2 1

	TIBLEŞ Mountains									TOROIAGA Mountains				
	BT-37	BT-30	BT-40	BT-15	BT-12	BT-8	BT-24	BT-7	BT-4	14	13	185	86	518
87Sr/86Sr		0.7065	0.7100	0.7073	0.7070	0.7067	0.7090	0.7088	0.7076					
SiO2	71.11	59.21	58.86	58.00	56.60	55.11	55.08	54.85	53.40	63.60	61.40	60.00	58.75	56.60
TiO2	0.19	0.30	0.50	0.60	0.80	0.90	0.65	0.70	0.55	0.57	0.62	0.75	0.80	0.86
Al2O3	16.42	18.22	16.34	15.29	19.53	14.04	18.32	16.03	17.02	16.82	16.52	16.57	16.62	15.21
Fe2O3	0.68	2.54	3.18	4.05	3.38	5.52	0.80	6.70	5.95	1.79	0.84	1.62	2.42	1.91
FeO	1.30	3.67	4.62	5.35	3.60	4.90	6.90	5.27	5.41	2.93	3.87	4.30	3.84	5.01
MnO	0.06	0.15	0.14	0.18	0.09	0.16	0.17	0.24	0.27	0.13	0.14	0.10	0.14	0.14
MgO	0.56	3.00	2.80	3.16	2.30	4.80	4.20	3.66	2.70	1.45	1.52	2.81	3.24	4.29
CaO	2.15	6.16	6.44	5.63	6.47	8.34	8.26	6.53	8.08	4.95	5.17	5.12	5.97	7.63
K2O	2.52	2.48	3.05	2.94	2.31	1.39	1.47	0.84	1.37	2.79	2.69	3.56	2.97	1.90
Na2O	3.16	2.60	2.69	2.70	2.64	2.77	2.91	2.62	2.70	3.31	3.38	3.49	3.21	2.86
P2O5	0.05	0.20	0.15	0.14	0.18	0.18	0.11	0.40	0.20	0.23	0.24	0.28	0.28	0.23
H2O+	1.47	0.68	0.84	0.97	1.08	1.21	0.64	1.56	1.50	1.67	1.42	1.37	0.87	2.05
S				0.65	0.11			0.27	0.17	0.06	0.78		0.02	0.56
Total	99.70	99.50	100.20	100.21	100.31	99.74	99.28	99.67	99.87	100.30	99.32	100.67	99.13	99.74
Sc		12	15	19	12	24	23	25	33	7	10	17	19	20
V	22	85	120	130	110	200	200	220	300	45	67	130	110	100
Cr		15	14	21	8	47	17	11	42	5	4	25	33	82
Co		13	12	16	9	20	17	16	24	10	8	13	22	13
Ni		8.5	12	17	6.5	50	17	13	39	4.5	7.00	7.5	13	18
Cu	25	33	20	75	67	65	50	34	105	13	12	16	15	16
Zn	46	160	220	230	55	190	110	190	140	67	54	46	90	48
Ga	22	15	17	15	20	15	18	15	16	23	22	22	23	12
Rb		100	80	10	95	65	100	30	75					
Sr	170	370	265	250	335	330	360	320	315	440	440	320	410	440
Y		20	22	24	20	20	19	23	21	25	28	28	24	23
Zr		170	185	240	165	145	110	140	100	340	260			115
Nb														
Ba	500	830	380	680	730	400	320	320	300	700	940	770	750	580
La										30	30	48	42	30
Pb	32	73	150	133	46	55	83	21	46	30	20	11	26	3.5
Yb		2.4	2.2	2.4	2	2	1.6	2	1.9	2.5	2.8	2.2	2.3	2
Th		14 2						5.6						
U		2 7						1.8						

Table 2 (continued).

	RODNA Mountains									BÂRGĂU Mts		CĂLIMANI Mountains								
	397	549	273	537	538	539	224A	545	544	C46	C44	1123	C13	C22	5036	1500	1492	5158	3377	1925
87Sr/86Sr	0.7070	0.7060	0.7040	0.7070			0.7030	0.7060					0.7090	0.7100	0.7100	0 7060	0.7030	0.7050	0 7070	0.7050
SiO2	73.00	65.30	64.50	62.20	60.50	57.70	55.70	55.70	50.50	53.59	51.28	69.37	63.57	61.48	61.23	57.10	54.65	54.21	53.00	52.01
TiO2	0.12	0.34	0.56	0.50	0.61	0.67	0.73	0.68	0.80	1.05	1.05	0.22	0.60	0.71	0.65	0.91	0.90	1.06	1.06	1.38
Al2O3	14.95	17.30	16.74	15.60	15.45	17.55	17.70	17.25	17.15	19.17	18.55	17.05	16.32	16.29	15.91	16.80	18.65	18.85	19.70	18.82
Fe2O3	1.32	1.87	1.30	3.17	3.20	3.56	4.08	2.82	4.50	9.00	9.72	1.31	4.80	5.97	3.39	3.65	3 88	5 42	3.24	4.59
FeO	0.43	1.79	2.17	2.25	2.37	2.84	3.37	3.51	4.20			0.34			2.28	3.68	4.24	2.43	3.58	4.68
MnO	0.04	0.11	0.15	0.11	0.12	0.14	0.14	0.10	0.15	0.17	0.17	0.03	0.10	0.12	0.12	0.14	0.17	0 08	0.12	0.16
MgO	0.20	1.44	1.45	2.36	2.34	3.43	4.52	4.13	6.46	3.91	5.31	0.35	3.89	3.39	3.14	3.54	4.44	2.69	3.76	3.67
CaO	2.23	5.17	5.89	3.95	5.18	6.05	8.14	7.90	9.55	8.68	9.88	2.58	5.08	5.52	5.64	7.10	8.02	7.71	8.34	8.53
K2O	2.77	0.96	1.40	3.14	2.82	2.13	0.49	1.64	1.30	0.83	0.61	3.18	2.24	3.12	2.74	1.94	0.77	1.96	1.71	1.10
Na2O	3.37	3.59	3.64	3.72	3.17	3.85	2.75	3.33	2.53	3.35	3.02	3.33	3.65	3.26	3.25	3.06	2.98	3.75	3.63	3.32
P2O5	0.16	0.26	0.24	0.58	0.26	0.40	0.00	0.14	0.49	0.15	0.12	0.15	0.18	0.22	0.43	0.19	0.26	0.37	0.41	0.30
H2O+	1.28	0.95	1.25	1.97	0.66	0.59	1.34	1.67	0.96			1.70			0.80	0.79	0.66	1.09	0.96	0.69
S	0.10	0.15	0.11	0.42	1.50	0.21	0.21	0.22	0.20			0.04			0.09	0.14	0.13	0.04	0.02	0.21
Total	100.20	100.11	100.36	100.17	99.77	100.29	99.65	99.85	99.90	99.90	99.71	99.65	100.42	100.08	99.67	99.94	100.40	99.66	99.53	99.78
Sc		6.5	3	12	14	13	26	25	26	27.7	33.3	3	13.5	17.1	22	16	18	24.5	25	13
V		48	27	105	130	155	220	230	270	251.9	247.1	5	90	134	145	130	150	220	235	160
Cr		10	6.5	82	80	46	85	68	220	2.5	48.5	2	160	62	70	34	43	43	57	45
Co		4	3 5	11	8.5	15	25	17	36			4			21.5	9	15	36	33	12
Ni		8	4.5	15	10	13	22	19	55	3.9	25	3	66.9	2.3	31	15	26	58	43	5.5
Cu	2	4	6.5	38	30	30	56	60	95	18.2	47 7	4	23.4	48.2	44.5	20	18	120	180	19
Zn	46	44	48	1230	70	78	44	72	46	92.5	80.2	60	54.1	67.3	50	55	65	63	47	74
Ga	11	15	11	18	20	20	18	20	15	18.9	17.7	22	17.2	18.7	26	13	13	32	31	17
Rb	125	50	65	115	110	75	30	55	40	20.0	38 58	110	79.04	133.6	110		60	90	60	60
Sr	220	270	180	280	330	380	350	310	1000	257.1	286.4	285	362.9	320.5	340	320	320	330	330	250
Y		20	11	21	22	20	28	13	23	26.4	24 8	8	16.89	26.23	38	26	24	39	37	15
Zr	38	95	65	145	180	140	85	100	80	113.8	89.07	175	141.1	213 8	270	140	100	185	195	52
Nb										6.44	5 05	14.21	10.51	12 09						
Ba	370	220	220	720	950	720	170	420	700	168.6	109.6	750	583.3	599.8	675	360	110	95	100	160
La				42	60					9.46	7 32		26.46	32 39	44	17		32	37	
Pb	13	2.5	5	230	13	8 5	6.5	7.5	8.5	7.72	3 12	27	10 66	17 08	45	12	4	20	26	2
Yb		1 8		2.4	2.5	2.3	2.2	1.8	1.8						2 6	2 1	1.7	1 9	2.4	1
Th		3.2		11.3	10.9	6.6	1 2		4 7	2.1	0.85	8.5	9.44	15.36	10 7	5 4	5 2	9 4	10 6	4.2
U		2		3 4	3 5	1.9	0 6		1.5			3.2	0	0	3 4	2 1	3 2	2.4	1.2	1

	GURGHIU Mountains										NORTH HARGHITA Mountains						
	G15	330	135	G7	G39	G124A	G32	183	G19	348	H28	H27	86	H33	826	H29	LV73
87Sr/86Sr		0 7030	0.7040	0 7036	0.7066	0 7043	0 7082	0.7040	0 7041	0 7040	0 7026	0.7055	0.7037	0.7068	0.7030	0.7061	0.7044
SiO2	62.49	61.73	61.31	59 52	58.78	58.06	57.87	57.00	55.59	50.99	69.93	65.88	64.70	60.37	59.86	57.18	56.10
TiO2	0 56	0 61	0 52	0 70	0 69	0.80	0.82	0.68	0.86	1.33	0.28	0.56	0.62	0.71	0.86	0.77	0.72
Al2O3	18.03	17 91	17.11	17 75	18.37	17.30	17.94	18.31	18.75	19.76	15.98	17.01	16.20	18.07	16.47	18.56	17.65
Fe2O3	5 37	4.26	2 07	3 40	6.39	3 18	6 84	7 84	5.22	4.68	3 15	4.56	3.83	5.74	2.46	7.09	2.39
FeO		1 41	3 32	2 84		3.60		0.56	2.36	4.89			0.93		2.76		4.22
MnO	0.11	0 11	0 12	0 15	0 14	0.13	0.13	0.16	0.15	0.17	0.10	0.08	0.07	0.12	0.14	0.12	0.18
MgO	2.43	1 94	3 78	3 00	3.01	3.63	3.62	2.26	3.64	3.92	0.34	0.79	2.48	2.94	3.00	3.28	4.41
CaO	6.20	5.55	6.24	6 14	6.92	7.08	7.18	6.61	7.46	9 32	2.82	3.79	5.85	6.36	7.70	7.40	7.34
K2O	1.70	1.23	1 21	1 40	1.57	2.25	1.44	1.85	1.27	1.14	2.83	2.26	2.92	2.09	2.25	1.51	1.35
Na2O	3.21	3.68	2 97	3.55	3.53	3.35	3.63	3.54	3.18	2.69	4.28	4.68	2.81	3.45	3.30	3.50	3.14
P2O5	0 14	0.27	0.20	0.13	0.15	0.11	0.19	0.26	0.17	0.34	0.11	0.22	0.20	0.13	0.10	0.14	0.20
H2O+		0.56	1.61	0.87		0.55		1.31	1.36	0.44			2.03		0.14		1.42
S		0.18	0.21	0 22		0.18		0.24	0.17	0.17			0.12		0.20		0.10
Total	100.25	99.54	100.80	99.67	99.55	100.22	99.65	100.72	100.18	100.06	99.82	99.83	99.76	99.97	100.09	99.56	100.02
Sc	16.8	8	9.5	15	17.6	13	21.3	8	16	19	2.3	8.1	7	16.6	14	20.3	20
V	106 4	70	90	160	136.3	140	162.6	80	160	220	8.2	33.1	82	100	105	153.2	90
Cr	30 2	3 5	45	5	14.1	8	29.8		8.5	5	3.6	9.1	54	13.3	15	7.8	21
Co		7	10	11		10		9	12	14			9		9.5		12
Ni	10.1		10	7	12	3	13.9	4.5	13	25	3.2	3.2	24	6	6.5	7.4	6
Cu	15 7	9	9.5	13	78.9	20	33	2.5	19	32	4 7	13.1	17	14.6	8	20.5	8
Zn	63.5	65	73	66	67.1	80	75	95	77	80	65.6	78.6		70.5	75	66.3	42
Ga	16.6	8 5	12	17	18.6	17	19.2	15	13	13	15.7	17.7	11	18.4	15	18.7	16
Rb	56 56	57 9	52	42	49.55	42 5	45.23	54	42	37	107.7	81.29	84	72.31	105	47.85	43
Sr	267.8	230	241	190	315 7	150	320	530	180	360	240.6	274.2	190	316.2	320	347.5	326
Y	18 1	14	20	23	21.34	17	23 41	25	20	20	14.85	24.82	12	21.73	17	22.91	12
Zr	125 9	95	115	140	153.2	120	150.6	220	125	75	217.5	211.2	125	156.5	120	125.9	160
Nb	8.4				8.23		9.9				8.98	12.41		9.22		7.05	
Ba	292.4	155	280	290	393.9	240	429.9	700	220	150	565.1	548	1015	388.1	340	320.5	210
La	17.98			30	22		26 74	50			28.35	35.11		20	32	16.31	
Pb	10.25		3 5		5.7	2	6.58	5.5	2.5		13.01	12.57		9.11	6	7.27	4
Yb			1.4	1.1		1.2		1.3	1.4	1.2					1.6		
Th	5.1				5 72		6 44				12.12	11 06		7 66		4.91	
U	0				0		0				0	0		0		0	

Table 2 (continued).

	SOUTH HARGHITA Mountains								
	CP/1P	H47	C82	837	C77	C76	H3	L18	H11
87Sr/86Sr	0 7060		0.7060	0.7050	0.7057	0.7057	0.7046	0.7080	0.7047
SiO2	65 98	64.72	62.40	61.00	60.50	59.50	57.92	57.42	54.90
TiO2	0.29	0.56	0.76	0.80	0.81	0.78	0.97	0.73	1.02
Al2O3	16.46	17.31	17.10	16.32	18 25	17 80	15.74	17.03	18.54
Fe2O3	1.30	3.96	3.09	2.38	2.85	4.03	4.89	2.03	7.59
FeO	0.85		1.31	3.03	2.56	1.96		3.98	
MnO	0.04	0.10	0 08	0.10	0.12	0.13	0.09	0.13	0.19
MgO	1.33	1.67	1.99	3.45	2.07	2.46	4.48	4.89	3.70
CaO	2.98	4.51	5.71	6.77	5.33	6.38	6.75	8.34	8.24
K2O	3.63	2.54	2.20	2.16	1.81	1.73	4.03	1.50	1.61
Na2O	4.60	4.03	3.81	2.83	3.92	4.04	3.91	2.98	3.72
P2O5	0.16	0.14	0.57	0.14	0.44	0.70	0.52	0.17	0.17
H2O+	0.70		0.78	0.90	1.36	0.28		0.95	
S	0.08		0.04	0.13	0.04	0.08		0.08	
Total	99.62	99.54	99.84	100.01	100.06	99.88	99.28	100.23	99.68
Sc	6.5	10.2	11	155	8	8.5	13.9	20	26.7
V	55	52.4	100	140	85	115	114.6	115	202.7
Cr	28	17.9	40	110	5.5	6.5	123.8	21	40.4
Co	8		7	11	7.5	8.5		8.5	
Ni	18	8.6	6	6	5.5	5.5	35.7	9	18.2
Cu	4	4.6	4	17	9	18	18.1	12	31
Zn	44	46.3	50	70	55	66	54	48	69.6
Ga	28	17.4	13	30	20	17	18.7	16	19
Rb	80	86.69	110	100	53	69	65.77	70	35.13
Sr	1450	529.3	440	370	770	820	2263.5	330	947.5
Y	18	17.01	14.5	15	14	16	18.47	22	25.26
Zr	190	138.7	150	190	115	115	234.7	160	107.8
Nb		16.59		25			18.73		11.79
Ba	900	771	560	480	950	700	2694.6	300	540.8
La	58	29.24	28		31	20	101.1		19.46
Pb	42	13.65	7.5	3	12	11	23.1	3.5	22.57
Yb			1.4	1.3	1.2	1.5		1.2	.
Th	12.4	11 48	11.4		11.2	8 9	14 04		5
U	3 8	0	2 8		2 6	2 2	0		0

were determined by nondispersive XRF spectrometry using a Si (Li) detector with 190 eV resolution at 5.9 Kev. The detection limits were 10 ppm.

$^{87}Sr/^{86}Sr$ isotope analyses were determined at the Institute of Nuclear Physics in Bucharest. The sample dissolution process used an admixture of HF and HNO_3 at 200°C for 24 hours. The dissolved Rb-Sr were separated using Amberlit (G Type II 200-400 mesh) cationic resin of analytical purity. The Sr separated from the column was nitrated and was isotopically analysed using a CH6 VARIAN MAT mass spectrometer, using thermal ionization.

The major and trace elements performed in the U.K. were determined on an automated Philips PW1400 XRF spectrometer. Sr isotope analyses were made using standard ion exchange separation technique and a VG 354 multicollector mass spectrometer at Royal Holloway, University of London. The determination made in Romania and the U.K. are comparable as can be seen in Tab. 2.

5. Petrochemistry

SiO_2-K_2O variations – ECVA samples mostly cover the medium-K calcalkaline field (Fig.2). High-K calcalkaline samples are mainly from the CGH segment and, to a lesser extent, from the OG and TTRB areas. A low-K signature characterizes some early volcanic products in the Călimani Mountains. Andesites and dacites prevail, followed by basaltic andesites. Basalts and rhyolites are sparse, the TTRB rhyolites being richer in K_2O than those of the OG and CGH sectors. The Călimani area covers the widest range of compositions (50-

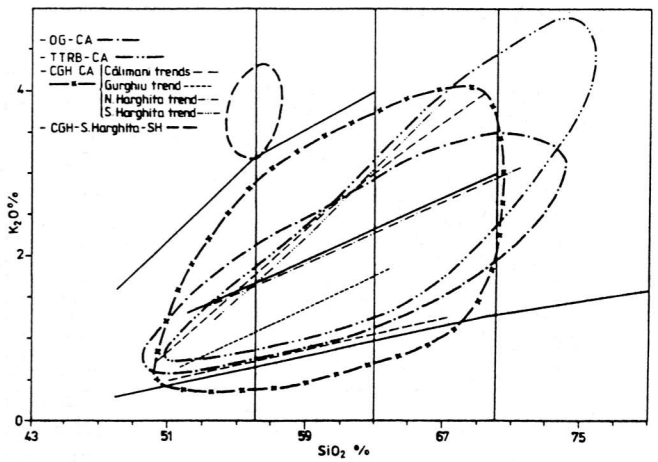

Fig. 2 – SiO_2 vs. K_2O diagram of Peccerillo & Taylor (1976b), showing fields for different ECVA segments.

71% SiO_2, 0.6-3.2% K_2O). In the southern Harghita, there is a clear trend of increasing K_2O and SiO_2 content along the arc. The southernmost centers lie in the field of shoshonites and banakites (Peccerillo & Taylor, 1976b).

Trace element behaviour – Sc and V are compatible, showing well-defined trends against fractionation indices, indicating fractionation of pyroxenes and amphiboles. The behaviour of Ni and Cr is less well-defined with some volcanic areas showing clear compatible behaviour (CGH segment), whilst other exhibit more scatter (OG and TTRB arc segments). Scatter may be due to magma mixing. Olivine and clinopyroxene have a major impact upon the Ni and Cr concentrations in a magma. Introduction of these minerals during an injection of basic magma into an evolved magma chamber may have created the scattered data. This can be backed up by petrographic evidence in some samples. Fractional crystallisation of olivine and clinopyroxene is indicated by subtraction of Ni and Cr. Y shows incompatible behaviour in many basalts and basaltic andesites, but is frequently compatible in more evolved lithologies, due to Y removal by amphibole. Sr is broadly compatible in many volcanic centers, but shows scatter against SiO_2, probably caused by variable plagioclase fractionation and accumulation.

Most other trace elements are incompatible. Many trends show scatter, but there is a lot of variation between volcanic centers. Crustal assimilation may cause some of the variability in incompatible element contents, particularly for Pb.

Sr/Ca-Ba/Ca or SB diagram – The SB diagram (Onuma et al., 1983) can be used to elucidate magma generation and evolution, based on the behaviour of Ca, Sr, and Ba. The distribution of these elements is influenced by magmatic processes (partial melting and fractional crystallisation) and reflects the nature of mantle-derived primary magmas and the interaction between mantle and crustal magmas (Fig. 3).

SB systematics in the ECVA is presented in Fig. 4, showing the evolution corresponding to each segment of the arc. The diagram reveals that trends start close to

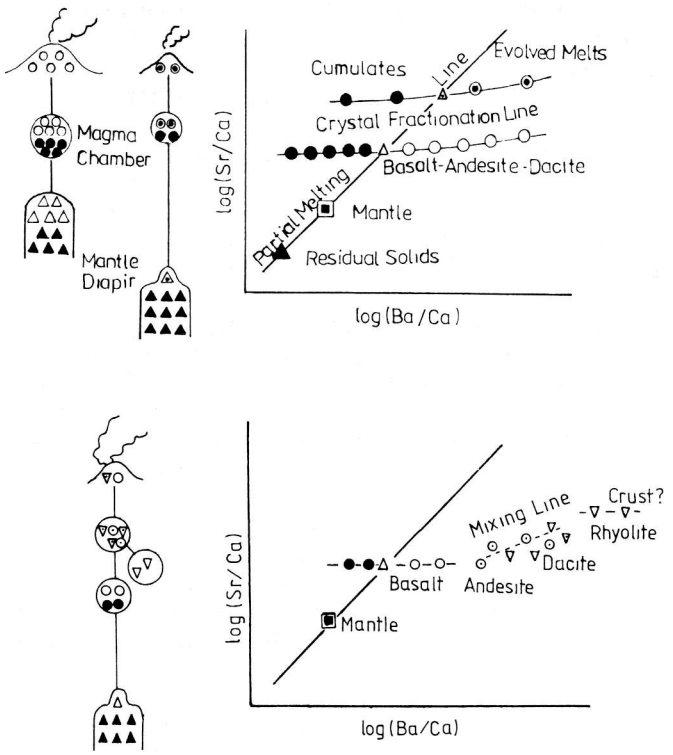

Fig. 3 – Schematic presentation of the relationship between magma genesis by partial melting in mantle diapirs and Sr/Ca and Ba/Ca ratios of generated primitive melts. The upper figure presents evolution related to different degrees of partial melting and consequent fractional crystallization. The lower figure shows evolution by mixing between mantle-derived melts and crustal melts. After Onuma et al. (1983) with modifications.

the PML only in the 2nd and 3rd stages of volcanic activity in Călimani. Close proximity to the Partial Melting Line (PML) suggests derivation of melts close to a chondritic mantle source. Rodna basalts and basaltic andesites are intermediate. The Oaș rhyolites are most distant from the PML suggesting a different source for these rocks, probably crustal. There is a specific Crustal Fractionation Line (CFL) trend for each area, with the exception of the Călimani, Toroiaga and South Harghita areas which show at least two parallel trends. The South Harghita volcanics evolved gradually from north to south to a lower degree of partial melting along the PML.

$^{87}Sr/^{86}Sr$ – Sr diagram – Separate diagrams are shown for the ECVA data obtained in Romania (partially presented by Peltz et al., 1987; Kovács et al., 1992) (Table 2) (Fig. 5a) and data obtained in the U.K. for the CGH arc segment (partially presented in Mason et al., in press) (Fig. 5b). The two data sets and the respective diagrams are similar. For almost all the areas, the rocks plot very close together between 200-400 ppm Sr, but with a large range of $^{87}Sr/^{86}Sr$ (0.7030-0.7110). South Harghita is the exception having a large variation in Sr content (300-2500 ppm) vs. an insignificant range of $^{87}Sr/^{86}Sr$ (0.7040-0.7050). Higher $^{87}Sr/^{86}Sr$ values are typical of acid rocks in the Oaș and Gutâi areas, but some high values were also found in the Călimani and Toroiaga areas.

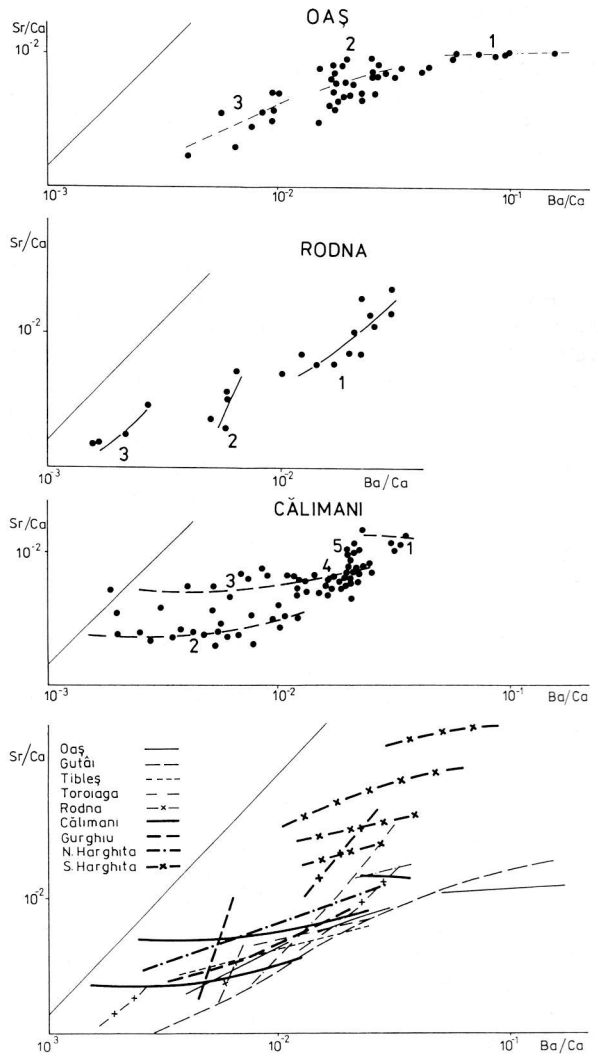

Fig. 4 – Sr/Ca-Ba/Ca systematics of magmatic rocks from Oaș, Rodna, Călimani and all ECVA areas. The numbers represent the space-time evolution (based on K-Ar age determinations and geological observations).

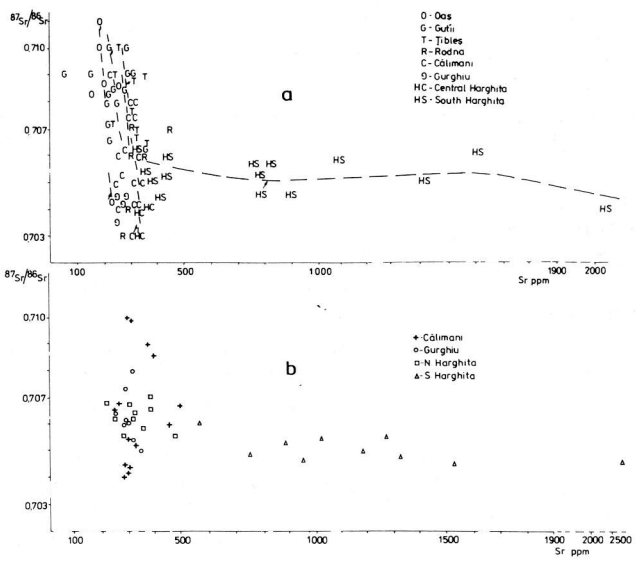

Fig. 5 – $^{87}Sr/^{86}Sr$ vs. Sr diagram for volcanics from all ECVA areas: (a) data from Peltz et al., 1986; Kovács et al., 1992 and Seghedi & Szakács, unpublished, and for volcanics of the CGH area alone (b) data from Mason et al., in press.

6. Discussion

Complex petrogenetic processes must be taken into account in order to identify and reconstruct the evolution of the ECVA magmas. Here we consider the dominant petrogenetic processes which include fractional crystallisation, magma mixing and assimilation, and attempt to identify the magma sources.

The magma source problem – It is difficult to envisage the source components of the ECVA without elimination of the later processes such as crystal fractionation, magma mixing and assimilation. None of the ECVA rocks, even the most basic ones, are representative of parental or close to parental mantle-source partial melts. Even the most depleted and uncontaminated rocks have undergone fractionation of mafic minerals (e.g. low Sc, Cr, Ni content corresponding to olivine and clinopyroxene fractionation). However, general trends suggest that most of the magmas are related to a mantle source (e.g. $^{87}Sr/^{86}Sr$-Sr, SB systematics). Using SB systematics, the basalts and/or basaltic andesites plot closer to the PML, but each area shows its own specific trend (Fig. 4). The only volcanics whose trend starts closer to the PML belong to the early stages of the Călimani area, suggesting a closer-to-chondritic composition for the mantle source of these rocks. Sr-Nd systematics also suggest that the most primitive magmas in the CGH arc segment are found in Călimani (Mason et al., in press). The other groups show a generally parallel trend with the CFL, starting at some distance from the PML, suggesting that the original mantle material was metasomatically enriched. Detailed trace element and isotope systematics seem to indicate that a second primary mantle reservoir existed for the South Harghita volcanics (Mason et al., 1995; Mason et al., in press). In most of the northern segments, but especially in the Oaș, Gutâi and Rodna areas, the youngest magmatic products are represented by basalts and basaltic andesites, whereas the older magmatic products are of acid composition (rhyolites in Oaș, Rodna and Călimani, dacites in Gutâi, Țibleș, Toroiaga and Călimani). These rocks plot further from the PML on the SB diagram. Being the earliest volcanic or subvolcanic products, it is very difficult to consider these rocks as derived from basaltic magmas through magmatic differentiation processes. It is more likely that these small volume acid magmas were partial melts of the lower crust. Crustal melting involves heating of the lower crust (close to the mantle-crust boundary) and could be facilitated by the intrusion of mantle-derived magmas from below. If the rhyolites could be entirely explained by this mechanism, then at least for part of the dacites, other contributing processes (e.g. magma mixing, assimilation) should also be envisaged. Another possible explanation for the first erupted acid products could be important fractional crystallisation of an andesitic mantle-derived magma which experienced important assimilation in the crust prior to eruption.

Degree of partial melting – Based on SB systematics, the degree of partial melting of primary basaltic magmas can also be deduced (Onuma et al., 1983). Different distributions along the PML are related to different degrees of partial melting. The approximate range of partial melting for the studied regions shows, is, with one exception (South Harghita), a relatively constant (10-15%) along the chain (Fig. 4). The South Harghita volcanics exhibit a progressively lower degree of partial melting (2-4%) southward along the chain (Szakács et al., 1993). Sometimes the same region may present different degrees of partial melting for different sequences of volcanic evolution, interpreted as discrete batches of magma supply (e.g. Călimani area).

Crystal fractionation – Compatible trace element behaviour (Ni, Sc, Co, Cr, V), incompatible element concentration (Rb, Th) and SB systematics support fractional crystallisation as the dominant petrogenetic process in ECVA magma evolution during storage in shallow magma chamber. Olivine and clinopyroxene and sometimes amphibole are the main mafic minerals involved in all areas except for the South Harghita where amphibole dominates over clinopyroxene and olivine as the fractionated phase (Mason et al., 1995). In the SB diagram (Fig. 4) in almost all the areas, the parallel distribution of the plots along the CFL is obvious only between the basaltic composition and the andesite-dacite boundary. Thereafter the plots are randomly distributed. This aspect possibly relates to the involvement of other processes such as magma mixing and/or assimilation.

Evidence for crustal contamination – The presence of crustal xenoliths is good evidence for crustal contamination. Isotopic studies suggest that crustal contamination of primary magmas is an important process, especially in Călimani and Gutâi areas (Peltz et al., 1987; Kovács et al., 1992; Mason et al., 1995; Mason et al., in press). This contamination directly correlates with the large volumes of extruded products (Fig. 1), suggesting either long storage in shallow magma chambers or the emplacement of sequences of magma batches that continuously allowed crustal heating and facilitated assimilation. Gurghiu and North Harghita volcanics are less affected by contamination and erupted less voluminous products. The high Sr isotopic ratio of the first erupted acid products from Oaș and Țibleș (0.710-0.711) could be related to an origin from a lower crustal source. Assimilation processes cannot be excluded, but may be less important. The earliest products from the Călimani area (Dragoiasa dacites) are similar and show high $^{87}Sr/^{86}Sr$ (0.710) and low $^{143}Nd/^{144}Nd$ (0.51245), suggesting a close relation to a crustal source. Their genesis as highly contaminated products of the depleted source of the Călimani volcanics seems more unlikely, although this is a possible alternative explanation. South Harghita volcanics have lower Sr isotope values, but modelling of combined assimilation and fractional crystallisation proves important contamination processes in the upper crust (Mason et al., in press).

Types of magma mixing – Magma mixing is another important process that influenced the evolution of ECVA magmas. Based mainly on the types of contrasting magma end-members and the extent of mixing, three kinds of magma mixing processes can be distinguished in the ECVA:

(1) mixing between mantle-derived and crustal-derived magmas in deep-seated magma chambers. This mixing was inferred from the SB systematics in the Oaș area (Fig. 4), where intermediary plots between rhyolites (which initiated magmatic activity from a crust-derived source) and basaltic andesites (youngest pro-

ducts and derived from a mantle-derived source) are randomly distributed, suggesting mixing between crustal- and mantle-type magmas. A very similar mechanism can be envisaged for the Rodna area where acidic rocks are older and basaltic ones are younger. This interpretation is also supported by the SB systematics (Fig. 4). The same mechanism, but to a lesser extent, also operated in the Gutâi area, where the older dacite products may represent small volume mixing products between crustal and mantle source melts. The dacites of Țibleș area would be interpreted in a similar manner;

(2) magma mixing between differentiated and unevolved mantle-derived melts in shallow magma chambers. This mechanism produced only minor volumes of products in the final stage of volcanism in the Gutâi and Călimani areas. The rocks contain mineral assemblages showing strong disequilibrium (commonly resorbed quartz, amphibole and biotite together with orthopyroxene, clinopyroxene and olivine). A similar mechanism but at greater depth can explain the generation of the two South Hargita shoshonitic domes, which contain crystals of Mg-olivine, clinopyroxene and corroded and/or resorbed quartz, amphibole and biotite. Cauliform crystal-clot inclusions with Hopper crystallisation which contain plagioclase + amphibole + pyroxene and glass are common in these rocks, and are characteristic of this type of mixing processes (Eichelberger, 1978);

(3) mixing between two slightly differentiated magmas. This kind of mixing is typical for some areas but does not strongly affect geochemical composition. Generally, the end-members are close in composition. We explain the random distribution of plots in SB diagrams by this mechanism. In the Țibleș, Rodna, Gurghiu and South Harghita areas, mixing between melts which have undergone different degrees of partial melting has taken place. This is supported by the parallel, but random distribution of points when compared with the PML (Fig. 4). However, the random distribution of the points along the CFL, in regions other than South Harghita could be related to the mixing of melts which had undergone different degrees of fractionation. Petrographic observations support this mainly in Gurghiu and South Harghita areas, where some disequilibrium mineral phases and/or different generations of the same mineral were recognized in the rocks. Cauliform inclusions are also present in these types of rocks, supporting the mixing processes hypothesis.

7. Concluding remarks

This general comparative approach to the generation and evolution of ECVA magmas reflects the complexity of petrogenetic processes, which can be summarised as follows: (1) the great bulk of the magma is mantle-derived, affected by a «subduction-related» geochemical signature. There are at least two mantle sources: one for the South Harghita volcanics and other for the remainder of the ECVA. This latter source is a complex one. It is most depleted and uncontaminated in the Călimani area and probably slightly metasomatised in the rest of the arc segments. An additional crustal-type source is supposed for the northern segments (OG, TTRB, Călimani), suggested by small volume acid magmas erupted at the inception of magmatic activity. The South Harghita source which shows lower Sr and Pb isotopic ratios, produced Sr and Ba-enriched magmas; (2) for most of the ECVA, almost constant degrees of partial melting of 10-15% are inferred, except for the South Harghita area where magmas result from gradually lower degrees of partial melting along the chain; (3) in areas with large volumes of erupted magmas (Călimani, Gutâi) petrogenetic processes are dominated by combined fractional crystallization and crustal assimilation. In areas with smaller volume erupted products (Oaș, Țibleș, Toroiaga, Rodna) extensive mixing between crustal- and mantle-source melts in deep-seated magma chambers, followed by fractional crystallisation and assimilation processes in shallow magma chambers is envisaged. The South Harghita volcanics suggest a complex combination of processes such as assimilation, fractional crystallisation and magma mixing, characteristic for each volcanic structure.

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