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Eruptive history and age of magmatic processes in the Călimani volcanic structure (Romania)

IOAN SEGHEDI[1], ALEXANDRU SZAKÁCS[1], ZOLTÁN PÉCSKAY[2] and PAUL R.D. MASON[3]

[1]Institute of Geodynamics, str. Jean-Luis Calderon 19–21, 70201 Bucharest, Romania; seghedi@geodin.ro
[2]Institute of Nuclear Research of the Hungarian Academy of Sciences, P.O. Box 51, Bem tér 18/c, H-4001 Debrecen, Hungary
[3]Vening Meinesz Research School of Geodynamics, Department of Earth Sciences, Utrecht University, Budapestlaan 4, 3584 CD Utrecht, The Netherlands

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Abstract: The Călimani Mountains represent the largest and most complex volcanic structure at the northern part of the Călimani–Gurghiu–Harghita range in Romania. Sixty-eight K-Ar ages (thirty-three new) provide constraints on the eruptive history of the Călimani volcanic structure between 11.3 and 6.7 Ma. The oldest rocks are from shallow exhumed intrusions, which pierced the basement between 11.3–9.4 Ma. The oldest stratovolcano was centered on the presently recognizable main volcanoes, Rusca-Tihu and the Călimani Caldera and grew very large (ca. 300 km^3), generating a large-volume (26 km^3) debris avalanche. Debris avalanche blocks dated between 10.2–7.8 Ma, suggest an edifice failure event at 8.0 ± 0.5 Ma. The Drăgoiasa Formation (9.3–8.4 Ma), Budacu Formation (9.0–8.5 Ma), Lomaş Formation (8.6 Ma), a number of Peripheral Domes (8.7–7.1 Ma) and Sărmaş basalts (8.5–8.3 Ma) were also active before the debris avalanche event. Volcanic activity continued from the Rusca-Tihu Volcano between 8.0–6.9 Ma, generating the "Rusca-Tihu Volcaniclastic Formation". The Călimani Caldera structure including pre-caldera and post-caldera stages was generated between 7.5–6.7 Ma, with an inferred collapse event at 7.1 ± 0.5 Ma. Monzodioritic-dioritic bodies in the central part of the caldera show ages between 8.8–7.3 Ma, implying several episodes of intrusions. Fractional crystallization was important in the generation of different magma series at lower crustal to shallow crustal depths, where plagioclase was the main crystallizing phase. Crustal assimilation affected most of the analysed samples to some degree through assimilation-fractional-crystallization (AFC) processes. Isotopic enrichment of the most basic rocks suggests that contamination processes affected the source of most parental magmas, except those of the Lomaş Formation. The initial stages of volcanism were most complex from the petrological point of view. The Drăgoiasa Formation (represented only by felsic rocks), for instance, suggests either fractionation from a basic parental magma and mixing with partial melts of (lower) crustal origin, or represents direct melting of the garnet bearing lower crust. The Lomaş Formation represents the most primitive magma, which reached the surface recording minimal interaction with crustal material and most closely characterizes the isotopic composition of the mantle source beneath the Călimani Volcano. The youngest volcanic rocks represented by the Călimani Caldera structure were derived from magmas that show a lower degree of partial melting and were largely affected by assimilation processes.

Key words: Eastern Carpathians, Călimani Mountains, petrology, volcanology, K-Ar data.

Introduction

The Călimani Mountains represent the northernmost and largest volcanic area amongst the 160 km long Călimani–Gurghiu–Harghita range in Romania (Fig. 1). The basement of this complex volcanic-magmatic centre is represented by: **1** — metamorphic rocks belonging to the Crystalline-Mesozoic zone of the Eastern Carpathians in the east, **2** — Cretaceous-Paleogene sediments in the north, pierced by a complex of shallow intrusions, belonging to the southern extension of the so-called "subvolcanic zone" of the Rodna-Bârgau area, and **3** — Neogene Molasse sediments of the Transylvanian Basin in the west. The largest and most prominent volcanic structure is the Călimani Caldera (Seghedi 1982, 1987). It is situated in the north of the area and covers almost one third of the Călimani Mountains.

K-Ar ages were previously published by Rădulescu et al. (1972), Peltz et al. (1987) and Pécskay et al. (1995). Petrological studies have been performed by Peltz et al. (1974, 1984), Seghedi (1987), Seghedi et al. (1995) and Mason et al. (1995, 1996).

The present study is based on 33 new K-Ar ages and reviews another 35 previously published ages produced in the same laboratory. Age data of Rădulescu et al. (1972) have not been used in this study. This new extended K-Ar data-base enables more constraints to be applied to the eruptive history of the volcanic edifice, as established by previous volcanological studies. An additional aim of this study is to discuss the petrological evolution of the area on the basis of existing geochemical data (Mason 1995; Mason et al. 1996).

Analytical methods

Rock samples dated in this study (Table 1) were taken from different types of magmatic rocks including those from lava flows, intrusions and blocks in volcaniclastic deposits. They were systematically collected using the methodology of Péc-

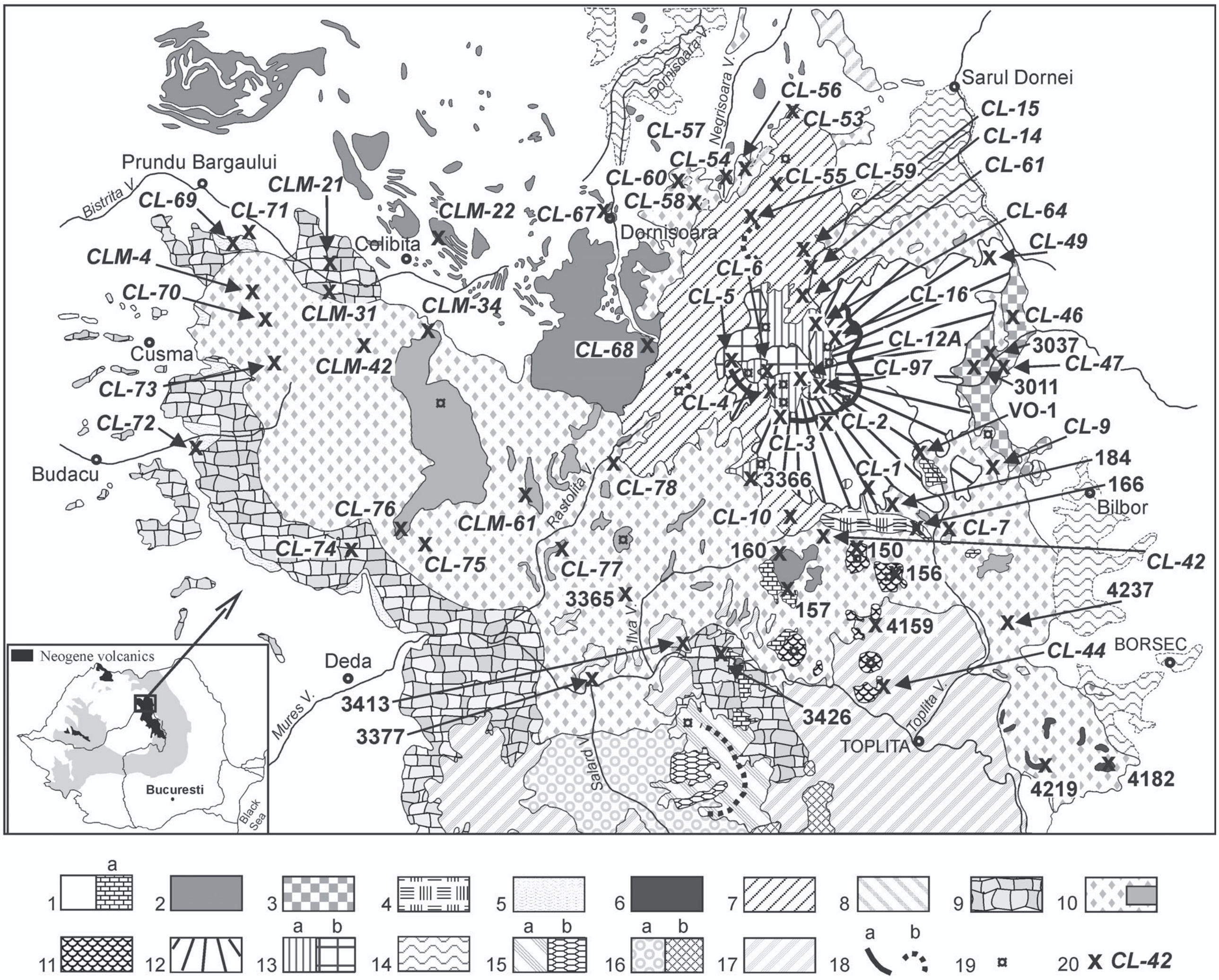

Fig. 1. Simplified volcanological sketch map of the Călimani and northernmost Gurghiu Mountains (according to Seghedi & Szakács in Szakács & Seghedi 1996) with location of sampling points for K-Ar determination. **1** — Pre-volcanic basement (a — in the interior of the volcanic area); **2** — Early intrusions (basaltic-andesites, andesites, microdiorites); **3** — Drăgoiasa Formation (dacites, rhyolites); **4** — Lomaş Formation (low-K andesites and dacites); **5** — Budacu Formation (andesites); **6** — Sărmaş basalt lavas; **7** — Rusca-Tihu stratovolcanic edifice (basaltic andesites and andesites); **8** — Aphyric andesite lavas; **9** — Rusca-Tihu debris avalanche deposit; **10** — Rusca-Tihu Volcaniclastic Formation with lava intercalations (andesites, basaltic andesites); **11** — Peripheral Domes (andesites and dacites); **12** — Călimani Caldera lava flows (andesites); **13** — Post-Călimani-Caldera rocks: a — monzodiorites, diorites, b — andesites, dacites; **14** — Upper Pliocene–Quaternary sedimentary basins; **15** — Jirca volcanic edifice: a — andesites, b — diorites; **16** — Fâncel-Lăpuşna pre-Caldera rocks: a — volcaniclastics, b — andesite and basaltic andesite lavas; **17** — Fâncel-Lăpuşna Volcaniclastic Formation (andesites, dacites); **18** — a — Caldera rim according to present topography, b — Crater rim according to present topography; **19** — Volcanic vent; **20** — Sample location.

skay et al. (1995). Rock samples were optically examined in thin sections and only the freshest or least altered material was prepared (crushed and sieved) for geochemical and geochronological study. One portion of the sieved fraction was used for the Ar analysis and another portion from the same sample was ground and used for K determination. All of the K-Ar work was carried out in the Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary, using the methods previously described by Pécskay et al. (1995). Further details of analytical methods and calculations of analytical errors are described in Balogh (1985). The decay constants used in the age calculations are given in Steiger & Jäger (1977).

Volcanic structure

The reconstruction of the volcanic structure of the Călimani Mountains has been carried out by reinterpreting previously published data (Peltz 1969; Teodoru et al. 1970; Seghedi 1982, 1987) using a set of 5 geological maps at scale of 1:50,000. Two of these have been published (Şarul Dornei, Poiana Stampei) whilst the other three (Bilbor, Negoiul Românesc, Topliţa) are as yet unpublished. A volcanological map of the Călimani volcanic area was first proposed in Szakács & Seghedi (1996) and is presented in Fig. 1.

The largest identifiable stratovolcanoes are represented by Rusca-Tihu and Pietrele Roşii and are built up mostly of basaltic andesites. They supplied huge volumes (~300 km^3 out of 420 km^3 for all Călimani Mountains volcanics) of volcanic deposits found all over the Călimani volcanic area on a surface larger then 900 km^2 (Szakács et al. 1997). Detailed volcanological survey supports the occurrence of large-volume debris avalanche deposits belonging to the Rusca-Tihu Volcano (26 km^3) (Szakács & Seghedi 1996, 2000) (Figs. 1, 3). This evidence implies that during its early history, the Rusca-Tihu Volcano (RTV) was much more imposing than today. Its height has been estimated at ca. 3000 m by Szakács & Seghedi (2000). Tectonic instability most likely led to edifice failure and the resulting debris avalanche reached distances of up to 55 km southwards and almost 40 km westwards. After this destructive episode, which would have resulted in a major topographic change, the volcanic activity continued from the central vents as well as from N–S-directed peripheral ones. The composition remained constant with the eruption of the same basaltic andesitic lavas, becoming more andesitic in the final stages. The post-debris avalanche proximal facies is represented by lava flows, in places autobrecciated, pyroclastic (mainly phreatomagmatic) flow and fall deposits and block-and-ash-flow deposits. The Rusca-Tihu stratovolcano then supplied debris flow deposits associated with hyperconcentrated flood-flow and normal stream flow deposits or lacustrine deposits (well exposed along the Mureş Valley) emplaced at intermediary to distal locations (Szakács & Seghedi 1996, 2000).

Besides the products of the Rusca-Tihu stratovolcano, other contemporaneous complex volcaniclastic formations, such as the Budacu Formation (at the western periphery), the Drăgoiasa Formation (eastern part) and the Lomaş Formation (southern-central part) have been identified. They are distinct from each another, according to their geochronological, petrographical, geochemical and volcanological features (Peltz et al. 1970, 1987; Peltz & Seghedi 1984; Seghedi 1987; Szakács & Seghedi 1996). Peripheral lava centres have been found all around the main volcanic edifices. The Drăgoiasa Formation is a dome-complex of aphyric dacites (occasionally rhyolites), associated with pyroclastic rocks in the east (Niţoi 1986). In the north and east, aphyric and normal andesitic lava vents are present (e.g. Măgura, Scaunul), whereas in the south basaltic-andesitic and dacitic lava domes prevail (e.g. Leul, Băieşul, Tarniţa, Mogoşul). They are referred to as Peripheral Domes (PD). In the south-easternmost part a large area of basalts (Sărmaş basalts — SB), which formed a shield volcano, are found and younger volcaniclastic deposits of basaltic-andesite composition cover them.

The Călimani Caldera structure represents the final major volcanic episode in this area. Its products, mainly lava flows, partially cover the series of NNE-trending older stratocones (Rusca-Tihu, Tămăul, Pietrele Roşii, Lucaciul) in the west, the Drăgoiasa dacite Formation, the volcaniclastic deposits of the older Rusca-Tihu Volcano and the Lomaş Formation in the east and south. The pre-caldera Călimani edifice consists of large-volume andesitic lavas, rich in silica and alkalies (Seghedi 1987; Mason et al. 1996). Flow directions of the lavas were dependent on local topography, with south and east-directed slopes, originating from at least four independent vents (Seghedi 1987). A huge volume of lava, estimated to be in excess of 10 km^3, was erupted in a relatively short interval of time, constrained by K-Ar data at ca. 300 ka (Pécskay et al. 1995). The actual caldera, with a summit rim altitude of ca. 2000 m a.s.l., was a result of the collapse initiated by the above mentioned effusive eruption. The horseshoe shape is assumed to be related to a half-block tilting downward the southeastern part from a NE–SW oriented hinge, resulting in a trap-door type caldera (Seghedi 1995). Post-caldera volcanism is represented by a few andesitic stratocones (e.g. Negoiul Românesc) in the interior of the caldera. A large monzodioritic-dioritic intrusion is exposed in an area of about 11 km^2 in the central part of the caldera. Dacitic domes located on the caldera rim (Pietricelul) and outer slopes (Drăguşul, Puturosul) are also post-caldera features. The central area has undergone extensive hydrothermal alteration (Teodoru & Teodoru 1966; Stanciu & Medeşan 1971a,b; Seghedi et al. 1985).

Discussion of the K-Ar ages and eruptive history

The new K-Ar age data are presented in Table 1. The volcanological sketch (Fig. 1) shows the sampling locations, including those of published ones, used in this study. The time distribution of the main age intervals of different volcanic and intrusive formations given by K-Ar data is summarized in Fig. 2.

The time span of the magmatic activity in the whole Călimani volcanic area is between 11.3 and 6.7 Ma. The oldest dated rocks are the exhumed subvolcanic intrusions of diverse composition — basaltic andesites, andesites and microdiorites, which pierced the metamorphic and Cretaceous-Paleogene sedimentary basement of the region (hereafter referred to as Early Intrusions — EI), confirming previous geological interpretations of their relative age (Török 1961). The ages of

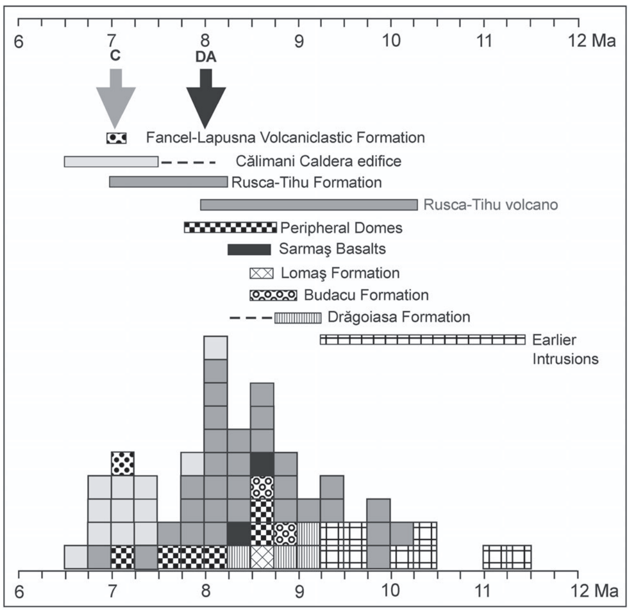

Fig. 2. K-Ar age histogram and time-space distribution of Neogene magmatic rocks in the Călimani Mountains (each block represents one sample). Abbreviations: **C** — Călimani Caldera-forming event; **DA** — Debris-avalanche event.

Table 1: Whole rock K-Ar ages for selected samples from Călimani Mountains volcanic area. The samples represent different formations or edifices generated during the evolution of the volcanic activity. Abbreviations: **vcl** — volcaniclastic deposit, **Aph** — aphyric, **B** — basalts, **BA** — basaltic andesite, **A** — andesite, **D** — dacite, **Mzd** — monzodiorite, **Py** — pyroxene, **Am** — amphibole, **Ga** — garnet, **Bi** — biotite.

No.	Lab. No.	Sample No.	Location	Rock type	Rock body	K (%)	^{40}Ar rad (%)	^{40}Ar rad (ccSTP/g)×10^{-7}	K-Ar age (Ma)
Early intrusions									
1	2879	CL-71	Pietroasa Valley	B-A	dyke	1.21	52.3	4.468	9.47±0.4
2	3762	CLM-22	Colibiţa Valley	Am-A	sill	0.88	9.0	3.299	9.65±1.0
3	2867	CL-54	12 Apostoli Valley	Am,Ga-A	dome	1.50	70.9	6.621	11.3±0.4
Drăgoiasa Formation									
4	2857	CL-46	Drăgoiasa Valley	D	lava	2.74	66.1	9.338	8.75±0.34
Budacu Formation									
5	3213	CL-69	Pietroasa Valley	Am-A	vcl. block	0.95	46.2	3.321	8.97±0.39
Sarmaş basalts									
6	3521	4182	Filpea Valley	B	lava	1.36	47.7	4.516	8.52±0.36
7	3522	4219	Cişcu Valley	B	lava	1.50	58.0	4.837	8.28±0.33
Peripheral Domes									
8	3520	4159	Zencani Peak	B	lava	1.63	49.1	5.120	8.06±0.34
Rusca-Tihu volcanic edifice									
9	2855	CL-60	Piatra Dornei Peak	Aph-A	lava	1.43	41.5	3.885	6.98±0.32
10	3765	CLM-42	Repedea Valley	Py-Ba	lava	0.99	37.4	2.958	7.68±0.37
11	2869	CL-55	12 Apostoli summit	Py-A	vcl. block	2.05	68.5	6.190	7.75±0.30
12	3761	CLM-21	Podişorenilor Hill	Py-AB	lava	1.35	50.5	4.181	7.98±0.34
13	2876	CL-59	Negrişoara Valley	BA	vcl. block	1.08	11.3	3.412	8.10±1.00
14	3523	4237	Hurdugaş Valley	BA	vcl. block	1.52	37.3	4.824	8.14±0.39
15	3764	CLM-34	Piatra lui Orban Peak	BA	lava	1.87	70.8	6.005	8.24±0.32
16	2858	CL-61	Haitei Valley	Py-A	lava	1.51	42.7	4.871	8.27±0,37
17	2880	CL-56	Ascuţit Peak	Aph-A	lava	1.80	35.0	5.885	8.39±0.41
18	3763	CLM-31	Şoimul de Jos Valley	Am-A	intrusion	1.08	79.6	3.579	8.48±0.32
19	2863	CL-53	Buza Şerbii-Pinţii Crest	Py-BA	lava	1.69	21.4	5.611	8.52±0.59
20	3766	CLM-61	Secu Valley	BA	lava	1.03	27.3	3.509	8.72±0.51
21	2859	CL-58	Prislop Valley	BA	lava	1.66	31.5	5.668	8.77±0.46
22	2875	CL78	Tihu Valley	Am-BA	dyke	1.40	50.2	4.808	8.80±0.37
23	2864	CL-64	Neagra Valley	Am-A	dyke	1.46	56.8	5.077	8.92±0.36
24	3214	CL-70	Pietroasa Crest	Py-AB	vcl. block	0.75	37.7	2.680	9.17±0.44
25	3217	CL77	Rastoliţa Valley	Py-A	lava	1.45	37.9	5.267	9.32±0.44
26	2999	CL-73	Bolovanul Valley	BA	vcl. block	0.86	54.4	3.149	9.35±0.38
27	3215	CL-74	Gălăoaia Mica Valley	Py-AB	lava?	0.74	17.5	2.806	9.76±0.79
28	3760	CLM-4	Pietroasa Valley	Py-AB	vcl. block	0.80	27.9	3.116	9.99±0.57
29	2661	VO-1	Voivodeasa Valley	B	lava	0.63	15.8	2.434	9.99±0.88
30	3216	CL-75	Gălăoaia Mica Valley	Py-A	lava?	0.81	27.6	3.212	10.17±0.58
Călimani Caldera									
31	2878	CL-49	Tomnatec Valley	Py-Am-A	lava	2.66	67.1	7.359	7.10±0.28
32	3976	CL-97	Călimani Quarry	Bi-Mzd	intrusion	3.72	58.8	10.052	7.26±0.29
33	3758	CL-12A	Călimani Quarry	Mzd	intrusion	2.19	39.5	6.834	8.02±0.37

various bodies belonging to this intrusive activity range between 11.3–9.4 Ma, covering a 2 million years time interval and coeval with the subvolcanic intrusions belonging to the Bârgău area (11.9–8.6 Ma) developed northward (Pécskay et al. 1995). They also crop out inside the volcanic area on the Zebrac Valley (10.1–10.6 Ma) or along the Mureş Valley in the Stânceni Quarry (9.5 Ma). In all these occurrences the cross-cutting relationships of the intrusive rocks with Miocene sedimentary strata are clear (e.g. Peltz et al. 1981).

As inferred from volcanological observations, the oldest stratovolcano was probably centered at the actual location of the main volcanoes — Rusca-Tihu and Călimani Caldera. This stratovolcano shows the largest age interval among all the volcanic structures of the Călimani Mountains (10.1–6.8 Ma). The most striking feature of the volcanic evolution of Rusca-Tihu is its synchronicity with the subvolcanic intrusions in the 10.1–9.1 Ma interval (Fig. 2), a critical time-period for the transition from intrusive to extrusive activity. The Rusca-Tihu Volcano, built up mostly of basaltic andesites, grew very large and voluminous between 10–7 Ma when it supplied a huge volume of volcaniclastic deposits, part of them related to a large debris avalanche event (Szakács & Seghedi 1996, 2000). The dating of the debris avalanche blocks gives an age interval between 10.2–7.8 Ma, similar to that found in the western side of the Gurghiu Mountains area (Seghedi et al. 2004). The youngest dated block in the debris avalanche suggests edifice failure of the Rusca-Tihu Volcano at ca. 8.0±0.5 Ma.

According to our volcanological field evidence, small-volume effusive and explosive volcanic activity was active in the surrounding area during the generation of the Rusca-Tihu Volcano. It produced the dacitic-rhyolitic Drăgoiasa Formation

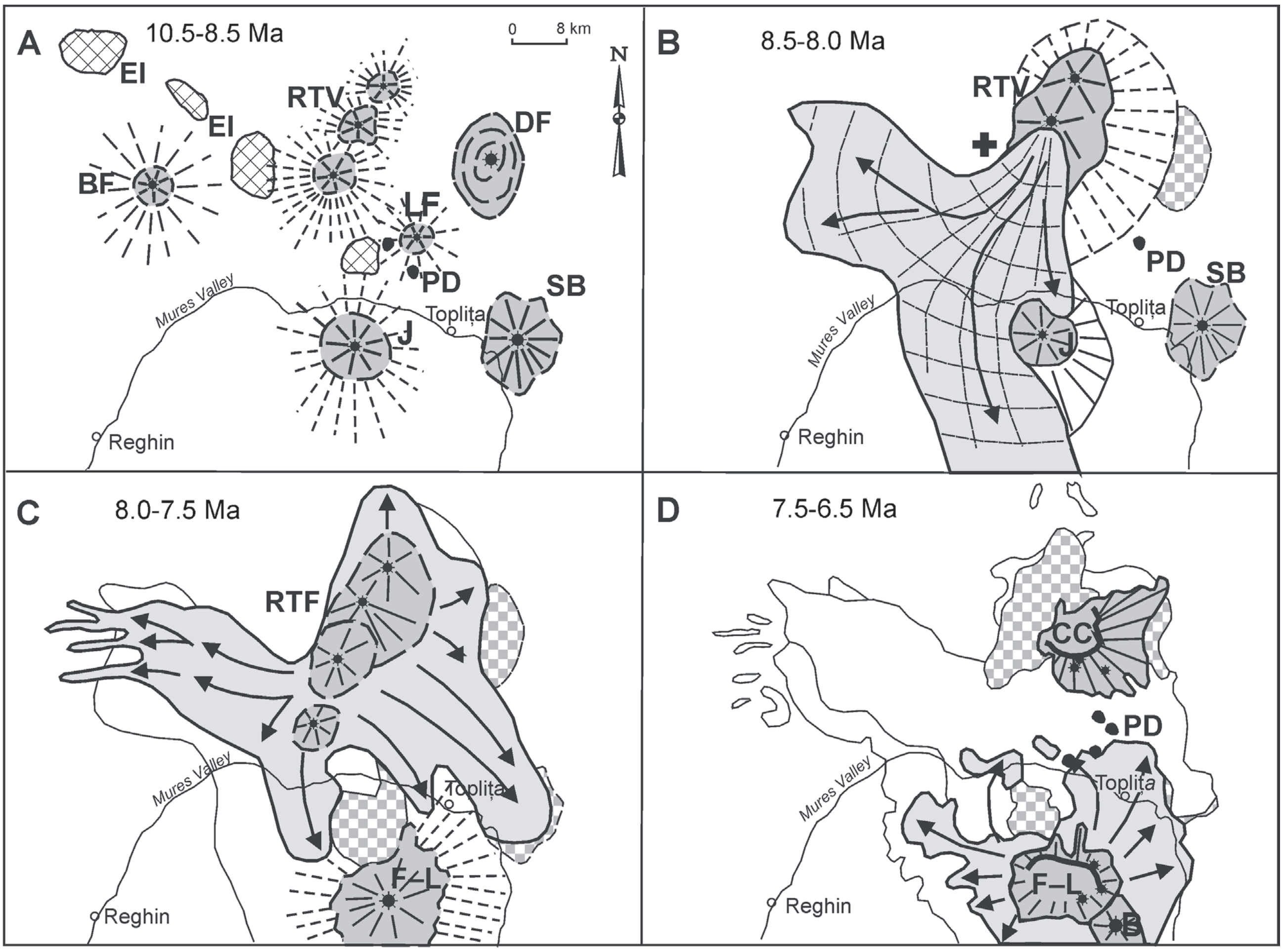

Fig. 3. Cartoons (**A, B, C, D**) showing the evolution of volcanism in the Călimani and northern Gurghiu Mountains (according to Szakács & Seghedi 1995, with modifications). Areas of active volcanic processes are represented by grey shadings for each time interval. **A.** EI — Early intrusions, DF — Drăgoiasa Formation, LF — Lomaş Formation, BF — Budacu Formation, RTV — Rusca-Tihu Volcano, SB — Sărmaş basalts, PD — Peripheral Domes, J — Jirca Volcano. **B.** Generation of Rusca-Tihu debris avalanche and volcanic activity at PD — Peripheral Domes, SB — Sărmaş basalts, J — Jirca Volcano. Thick cross in B cartoon indicates tectonic uplift. Arrows show assumed dispersion path directions of volcaniclastics. **C.** RTF — Rusca-Tihu Volcaniclastic Formation, FL — Fâncel-Lăpuşna Volcano. Arrows show assumed dispersion path directions of volcaniclastics. **D.** CC — Călimani Caldera Volcano, PD — Peripheral Domes, FL — Fâncel-Lăpuşna Volcaniclastic Formation, B — Bacta Dome Complex.

(9.3–8.4 Ma), andesitic-dacitic Budacu Formation (9.0–8.5 Ma), low-K andesitic-dacitic Lomaş Formation (8.6 Ma), andesitic-dacitic Peripheral Domes (8.7–7.1 Ma) and Sărmaş basalts (8.7–8.3 Ma). All these peripheral volcanic centres were active before the main debris avalanche event (Fig. 3).

Following the inferred debris avalanche event (at ca. 8 Ma), the volcanic activity continued from the same Rusca-Tihu Volcano, as well as other peripheral vents during an interval between 8.0–6.8 Ma. The deposits of post-debris-avalanche volcanic activity have been denominated as the "Rusca-Tihu Volcaniclastic Formation" — RTVF (Szakács & Seghedi 1996) and RTF hereafter (Fig. 3). On the basis of the K-Ar data presented here we have split the RTV and the RTF units. Basaltic andesites and other andesites were generated in this interval and a peripheral volcaniclastic apron was constructed consisting of a complex lithological assemblage of proximal pyroclastics (of both fall and flow origin) with several intercalated lavas, and secondary reworked sequences (debris flow, hyperconcentrated flood flow and fluvio-lacustrine deposits). Depositional environments ranged from terrestrial to lacustrine. The Călimani Caldera is the youngest and most important post-debris-avalanche volcanic feature and partially covers a series of NNE trending older stratocones of the RTV, toward the west. The pre-caldera volcanic rocks (PC) have been dated between 7.1–6.8 Ma, while volcanologically recognized post-caldera volcanic events (CP) suggest a similar age interval between 7.3–6.7 Ma (Fig. 3). The short time-interval of pre- and post-caldera evolution is notable (several hundred thousand years). However, the monzodioritic-dioritic intrusion exposed in the central part of the caldera shows a larger age interval, between 8.0–7.3 Ma, which in part overlaps with the pre- and post-caldera volcanic events (Fig. 2). Taking into account the youngest age of pre-caldera lava flows we can infer the caldera collapse event around 7.1 ± 0.5 Ma.

One sample of an amphibole-pyroxene-bearing andesite, collected from volcaniclastic deposits attributed to the Fâncel-Lăpuşna Volcaniclastic Formation — FLVF (Szakács & Seghedi 1996) yields an age of 7.1 Ma, which is in the same range with dated samples of this formation from the northern Gurghiu Mountains, which designate the moment of Fâncel-Lăpuşna Caldera generation, as a consequence of a major Plinian eruption (Szakács & Seghedi 1996; Seghedi et al. 2004). Caldera-type edifices Călimani and Fâncel-Lăpuşna are the most important of the Călimani–Gurghiu–Harghita range, being generated in a short time interval at around 7 Ma, almost contemporaneously (Fig. 3).

Petrological evolution in the light of K-Ar dating

This section discusses the geochemical and petrological evolution of the distinct volcanic formation identified using K-Ar geochronology. We use the geochemical data-base from Mason, (1995) consisting of 67 rock samples, most of them collected from the same outcrops as the K-Ar samples used for this study. Seghedi (1987), Mason (1995) and Mason et al. (1996) already pointed out that the Călimani volcanic area is petrogenetically very complex and is a result of various contributions of mantle and crust materials in the genesis of primary mafic magmas. In spite of the fact that the area displays huge volumes of basalt and basaltic andesite, the low-K dacites belonging to the Lomaş Formation have been found as the most isotopically primitive (Mason et al. 1996). The Călimani basalts in general show low MgO (<7 wt. %), Ni (<70 ppm) and Cr (<210 ppm), suggesting fractionation of mafic phases such as olivine and clinopyroxene during magma ascent.

The TAS (total alkali vs. silica) diagram (Fig. 4) indicates a broad range of rocks from basalt to rhyolite, dacites and rhyolites are specific for Drăgoiasa Formation and Lomaş Formation (almost exclusive) and some post-caldera rocks. EI, RTV and RTF volcanic products range from basalts to andesites. Pre- and post-caldera rocks are mostly andesitic, but with a slightly higher alkali content in Budacu Formation and Peripheral Domes rocks, which plot in the andesitic field. SB rocks have a slightly elevated alkali content. Incompatible element abundances of Călimani calc-alkaline basalts normalized to primitive mantle (Fig. 5) are variably enriched in large ion lithophile elements (LILE) and light rare earth elements (LREE) and also show variable Nb depletion, a characteristic feature for subduction-related magmas. The SB basalts are most enriched and show a negative spike of Sr as compared to other basalts.

Correlation of $^{87}Sr/^{86}Sr$ with SiO_2 provides evidence for the occurrence of both source contamination and assimilation in the volcanic suites of the Călimani region (Fig. 6). The fractionating mineral assemblage (i.e. plagioclase, olivine and pyroxenes) mainly caused the increase in SiO_2, whereas the shift toward higher $^{87}Sr/^{86}Sr$ ratios is related to assimilation. Source contamination is linked to the increasing $^{87}Sr/^{86}Sr$ ratio of most primitive rocks (basalts and basaltic andesites). The large range of geochemical and isotopic characteristics observed between rocks of the different volcanic formations may have resulted as a consequence of magma evolution at multiple locations in small-volume pockets. These magmas evolved independently from each other on the way to the surface. The Lomaş Formation dacites show minimal interaction with crustal material and may represent the evolved composition of a primitive magma, which did not reach the surface, but which is isotopically closest to the mantle source. The plot of the most primitive RTV, RTF and SB (along with an inferred composition for the Lomaş Formation group) suggests variable source contamination. Increasing $^{87}Sr/^{86}Sr$ ratios from this source contamination trend may be related to variable crustal contamination. The progressive increase of $^{87}Sr/^{86}Sr$

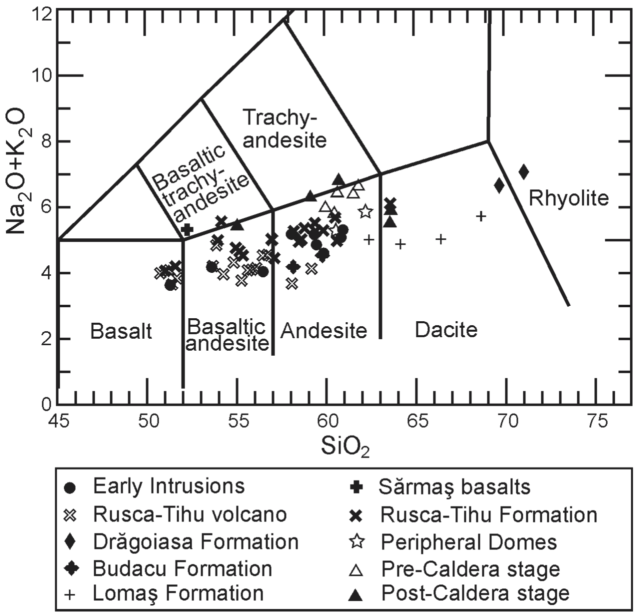

Fig. 4. TAS diagram for Călimani Mountain samples. Symbols and abbreviations as in Fig. 3. Further symbols: **PC** — pre-caldera stage, **CP** — post-caldera stage. Data from Mason (1995).

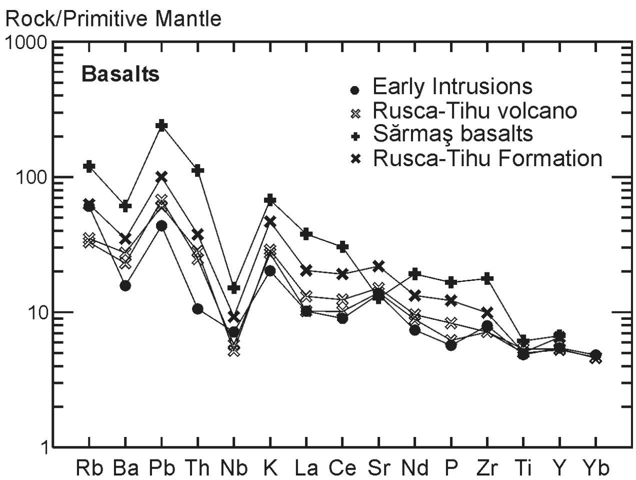

Fig. 5. Primitive mantle normalized incompatible trace element diagrams for calc-alkaline basalts from Călimani Mountains, using the normalizing coefficient of Sun & McDonough (1989). Data from Mason (1995).

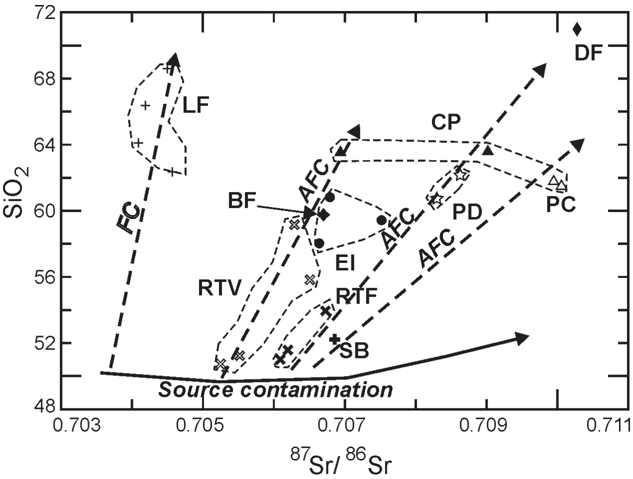

Fig. 6. $^{87}Sr/^{86}Sr$ vs. SiO_2 diagram for Călimani Mountains samples. Symbols as in Fig. 4. Symbols and abbreviations as in Figs. 3 and 4. Data from Mason (1995).

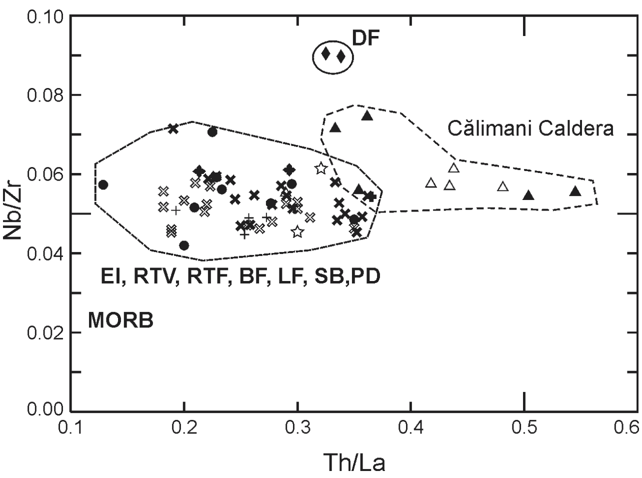

Fig. 8. Nb/Zr vs. Th/La diagram for Călimani Mountains samples. Symbols as in Fig. 4. Data from Mason (1995). Symbols and abbreviations as in Figs. 3 and 4. Data from Mason (1995).

ratios for similar SiO_2, may suggest increasing assimilation, along with fractional crystallization trend (FC), of successive magma batches, in the evolution of Călimani Caldera. The highest $^{87}Sr/^{86}Sr$ ratios and SiO_2 of Drăgoiasa Formation rocks indicate a strong crustal influence. Since these rocks also show HREE depletion (Mason 1995) indicating the direct implication of garnet in their genesis, it is likely that their parental magma may have mixed with, or represent partial melts of a garnet-bearing lower crust. The distinction between source contamination and crustal assimilation can be more easily recognized using Sr-Nd and O isotopic modelling (James 1981; Ellam & Harmon 1990), since oxygen isotope enrichment is a sensitive indicator of crustal contamination (Mason et al. 1996) (Fig. 7). Assimilation-fractional crystallization (AFC) curves have been modelled using the most isotopically primitive compositions, which belong to samples C65 (RTV) and C10 (EI) and a crustal assimilant, represented by the average value for Eastern Carpathians local crust (Mason et al. 1996). Between 5 and 20 % upper crustal contaminant is required in the AFC modelling (EI, RTF, Drăgoiasa Formation, pre-caldera volcanic rocks). Up to 2 % source contamination can explain the $^{87}Sr/^{86}Sr$ variability of the basaltic composition belonging to various formations. Lomaş Formation dacites are the most unaffected by source contamination. These variable source contamination processes suggested for the Călimani magmas are perhaps related to differences in the mantle wedge composition affected by variable sediment and fluid addition. Extreme $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ values of the Drăgoiasa Formation rocks suggest a crustal origin of their magmas is most likely.

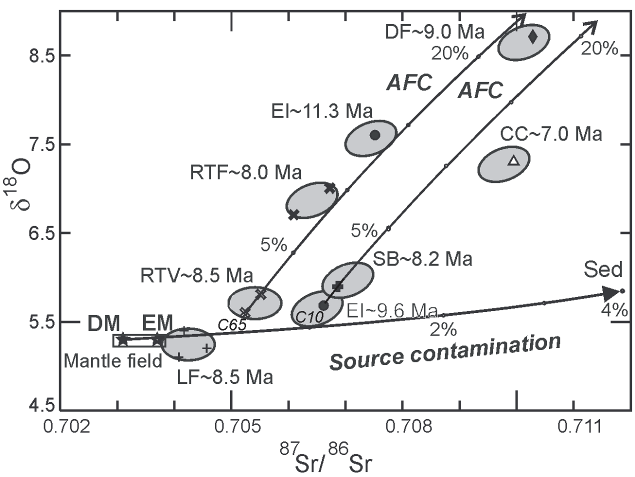

Fig. 7. $^{87}Sr/^{86}Sr$ vs. $\delta^{18}O$ variation for Călimani Mountains samples. Assimilation-fractional crystallization (AFC) and bulk mixing models intend to explain Sr and O isotope variation of Călimani magmas. For AFC: r = 0.4 trend (degree of assimilation/degree of fractionation) is shown with tick marks for every 5 % of consumed crust. D_{Sr} = 1.2 in all calculations. Symbols and abbreviations as in Figs. 3 and 4. Data from Mason (1995).

The ratio of high field strength elements (HFSE) such as Nb and Zr, can provide insight into variations in magma source composition (e.g. Davidson 1996; Singer et al. 1996). Nb and Zr are depleted in subduction-related magmas and are assumed to be dominantly mantle-derived, being relatively immobile under hydrothermal conditions. However, a small degree of modification to these ratios may be possible during bulk crustal assimilation, to lower Nb/Zr ratios (crustal Nb/Zr is ~0.06 — Mason 1995) corresponding to higher or lower Th/La (crustal Th/La is ~0.36 — Mason 1995). The Nb/Zr-Th/La diagram (Fig. 8) shows Nb/Zr values starting from ~0.05 (close to a typical MORB value). Higher Nb/Zr ratio characterizes only the Drăgoiasa Formation rocks. However, in the same range of Nb/Zr, Th/La is much higher for the Călimani Caldera rocks. This suggests either lower-degree partial melting of a similar source, or crustal assimilation, or both. If crustal assimilation did play a role (as suggested by high $^{87}Sr/^{86}Sr$) then the local assimilated crustal component has not yet been identified in this area (highest crustal Th/La = 0.5; Mason 1995). Co-variance of Ni with Rb (Fig. 9) is considered to result from partial melting along the Rb trend and fractional

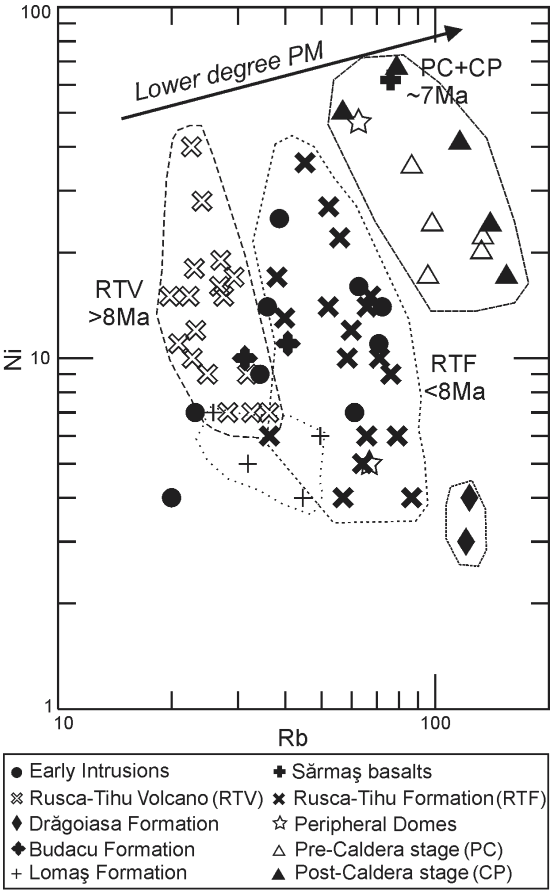

Fig. 9. Ni vs. Rb diagram for Călimani Mountains samples. Symbols as in Fig. 4. Symbols and abbreviations as in Figs. 3 and 4. Data from Mason (1995).

crystallization along the Ni trend. As the highest Ni content of Călimani basalts does not exceed 60 ppm, it is clear that all the magmas were affected by fractionation before rising to the surface and are far from primary melts in their composition. Crystal fractionation most likely started to occur at the deep levels, maybe at the crust — mantle boundary. Rb variation, however, suggests higher degrees of partial melting for EI (11.5–9.5 Ma), RTV, Lomaş Formation and most of the volcanic activity between 10–8 Ma. Călimani Caldera resulted from magmas, which for the whole Călimani Mountains volcanic structure resulted from a lower degree of partial melting, reflecting a general decrease of partial melting through time. This may imply a temperature decrease in the mantle-source toward the end of volcanic activity in the Călimani volcanic area, but in parallel it suggests a temperature increase in the upper crustal magma chamber (i.e. Călimani Caldera), as result of the ascent of successive magma batches, significant for increased assimilation along with fractionation.

Conclusions

The Călimani volcanic area was active between 11.3 and 6.7 Ma, with a complex magmatic history involving eruptions from multiple vents. The earliest volcanic activity developed between 10.5–9.5 Ma, partially overlapping previous intrusive magmatism. We suggest the following scenario for the evolution of this volcanic structure:

1 — Shallow intrusions pierced along a NNW–SSE trend the metamorphic and Cretaceous-Paleogene sedimentary basement rocks of the region between 11.3–9.4 Ma, representing the southern extension of the intrusive activity in the Bârgau area (11.9–8.6 Ma) in the north. Enriched mantle source, fractionation and assimilation at crustal levels were important petrogenetic processes in the generation for these rocks.

2 — Several major volcanic formations were formed throughout the whole Călimani area during the initial stages of the extrusive activity between ~10–8 Ma: Rusca-Tihu Volcano (which covers the entire interval), Drăgoiasa Formation (9.3–8.4 Ma), Budacu Formation (9.0–8.5 Ma), Lomaş Formation (8.6 Ma), Peripheral Domes (8.7–7.1) and Sărmaş basalts (8.7–8.3). The main stratovolcano, centered on the actual location of the Rusca-Tihu and Călimani Caldera structures was the largest and most voluminous. Its instability led to a large debris avalanche event at 8.0±0.5 Ma. The magmatic activity at this time was complex, with different volcanic formations resulting as a consequence of magma evolution at multiple locations. Drăgoiasa Formation rocks either fractionated from a more basic, but enriched parental magma, and mixed with partial melts from a garnet-bearing lower crust, or represent direct partial melts of a garnet-bearing lower crust. Lomaş Formation magmas fractionated in deep-seated magma chambers, but ascended to the surface with minimal interaction with crustal material. The Rusca-Tihu Volcano suite is the most representative for the Călimani area, suggesting both source contamination and AFC processes, in shallower larger-volume magma chambers. The Budacu Formation magmas are similar to the Rusca-Tihu magmas, but are more fractionated. The Sărmaş basalts magmas experienced the largest source contamination.

3 — Following the debris avalanche event, between ~8–7.5 Ma volcanic activity continued at the same Rusca-Tihu Volcano and other peripheral vents, generating the RTF Formation. These magmas represent a new batch and exhibit a larger degree of source contamination, lower degree of partial melting and AFC processes in shallow magma chambers. The magmatic activity at this time tends to concentrate in one major volcano as a consequence of magma system evolution towards smaller number of locations.

4 — The Călimani Caldera was generated between ~7.5–6.7 Ma (collapse at 7.1±0.5 Ma) and is the most important post-debris avalanche volcanic structure. Pre-caldera volcanic products show ages between 7.1–6.8 Ma, while post-caldera volcanic events are dated between 7.3–6.7 Ma. A post-caldera monzodioritic-dioritic intrusion exposed in the central part of the caldera shows a larger age interval, between 8.8–7.3 Ma and, according to field evidence, was probably emplaced in several intrusive episodes. Some Peripheral Domes were generated in this interval. These rocks are envisaged as a new batch of magma giving the pre- and post-caldera stages, as well as some of the Peripheral Domes. This magma fractionated from a source characterized by more significant enrichment, evolved AFC and lower degree of partial melting, as compared with earlier magmas. This implies lower temperatures at the source and higher temperatures in a unique upper crustal magma chamber, at the termination of volcanic activity.

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