Closure report on the no. K 115472 NKFIH-OTKA project

Eruptive rates and erosional dynamics of the Inner Carpathian volcanism as constrained by volumetry, radiometric chronology, and paleomagnetism

1 Introduction

During the years 2015-2019, a complex volcanological / volcanic geomorphological project has been conducted on the Carpathian Miocene to Quaternary volcanism. Main objectives of the research, as listed in the project proposal, included obtaining (1) main time intervals of volcanism, (2) periodicity in volcanic output, (3) periods of forming volcanic mountains, (4) erosional dynamics, (4) paleogeographic evolution related to volcanic activity, and (5) Quaternary tephrochronology. These objectives, with a main concept to address the time-space evolution of the whole Carpatho-Pannonian region, were planned to be achieved in selected study areas such as: the Mátra Mts; the forelands of Mátra and Bükk Mts; the Börzsöny-Visegrád Mts; and the Ciomadul (Csomád) volcano. During the project, in addition to the the study areas named above, the East Carpathian volcanic range was also succesfully selected for study, which well represents a peculiar time-space evolution within the Miocene to Quaternary volcanism.

One of the main objectives was calculating eruptive (magma output) rates. In full, among other details of volcanic activity, this was possible and has been completed for Ciomadul volcano (published papers) and the East Carpathians (submitted paper); and is in progress for the Börzsöny Mts. (conference presentations and manuscript in preparation; for details of all of them, see point 4). However, such an approach could not have been completed for the pyroclastic successions of the Mátra Mts. and the Bükk forelands. In these areas, the stratigraphic units have been turned out to be much more complex and unclear than expected in the project proposal. Thus, exact volumes of stratigraphic units could not have been computed (not to mention that a large amount of them is buried due to basin subsidence). However, to be documented in the following (see point 4), we have made significant steps toward clarifying the stratigraphy, by identifying pyroclastic-fall marker horizons, and by ignimbrite correlation (e.g. Mátrabérc, Tar, Bükk foreland ignimbrites). In other words, although in these areas the results were not sufficient for volume and thus magma output rate calculations, they hopefully pave the way toward a significantly improved volcanic stratigraphy, which is the focus of a new NKFIH-OTKA project (no. K 131894).

This project closure report will consist of the following parts: (2) realization of the project in time, (3) summary of publication activity and conference attendances, and (4) a detailed summary of obtained results by study area.

2 Realization of the project in time

In the yearly reports, full details of achievements have been given on field work, meetings, conference attendances, including participant names and timing. Hereby we give an account on these activities financial year by year (i.e. 01 Sept to 31 August 2015-2019), completed by data about minor changes relative to project participants and project budget plan.

2015/16: The activities in the first year included a number of field work to the target areas (mostly Ciomadul, Börzsöny, Mátra Mts. and Bükk southern foreland) with volcanological mapping and rock sampling for geochemistry, K-Ar and paleomagnetic measurements as defined in the project plan. It should be mentioned that several of the time-consuming paleomagnetic and K-Ar measurements (field and lab work), along with students' employement, were re-scheduled and put earlier relative to the plan, which resulted in an increase of personal costs as it appears from the approved re-arrangement of budget plan. Most important meetings of project participants included the XII Székely Geological Meeting, Cristuru Secuiesc/Székelykeresztúr, the 8th Hungarian Geographical Meeting, and EGU General Assembly, Vienna (first results on Ciomadul); and first visit to GEOPS, Orsay, initiating K-Ar dating of Börzsöny Mts. Moreover, visit to Pisa and Fuerteventura to give presentations and consult with international experts were also helpful in order to better constrain the project objectives. By contrast, visit to Beregovo (Beregszász), and Lvov (Lemberg), Ukraine, to initiate field work in the Northeast Carpathians, failed in the following years partly for political and economic reasons.

2016/17: Extensive field work continued in the financial year mostly at Ciomadul, the East Carpathians as a whole, the Mátra, Cserhát and Bükk foreland, again with the main goal of rock sampling. More field work and participation of more meetings than planned resulted in a slightly increased personnel cost per diem (approved). Most important meetings of participants included the 2nd European Mineralogical Congress, Rimini; EGU General Assembly, Vienna; and paleontological meetings in Tata-Tardos and München; where mainly the results on the Miocene volcanic and interbedded sedimentary successions were presented. Moreover, an organized session at the Hungarian Academy of Sciences (MTA) about new OTKA-funded results comprised four talks by project participants devoted to the research at Ciomadul and in the Bükk foreland. Unfortunately, senior participant Kadosa Balogh deceased during the year, and a new senior participant Balázs Székely joined the project, which implied a partial rearrangement of tasks (principal investigator D. Karátson undertook systematizing previous K-Ar datings, whereas B. Székely took part in arranging GIS studies).

2017/18: Additional samples were collected this year (e.g. Bükk foreland, High Börzsöny), and the main emphasis was put on presenting the results at international forums and writing papers. Most significant meetings were as follows: 8th Petrological and Geochemical Meeting, Szihalom, oral presentations and field-trip leading (on the Bükk foreland); international volcanological workshop at Ustica, oral presentations, and INTAV meeting, Moiceu de Sus, keynote talk and field-trip leading (on Ciomadul tephrochronology); 4th Central European Geomorphology Conference, Bayreuth, oral presentation (on Ciomadul volcanic geomorphology); EGU General Assembly, Vienna, oral presentation (on correlation of Mátra and Bükk foreland ignimbrites). Attendance of more conferences than originally planned justifies extra foreign travel and conference costs (excluding per diem).

2018/19: Additional, minor sampling and field checking were conducted in Hungarian study areas, including paleontologically important sites, and the main activity was to finish a number of scientific publications. Outstanding meetings included 10th Cities on Volcanoes, Naples, and XX Székely Geological Meeting, Sfintu Gheorge/Sepsiszentgyörgy, oral presentation (on Ciomadul); again an organized session at Hungarian Academy of Sciences (comprising four presentations on the Bükk foreland and High Börzsöny); 21st Mining Metallurgy and Geology Conference, Baia Mare/Nagybánya, and 25th European Current Research on Fluid and Melt Inclusions, Budapest, oral presentation (on the Bükk foreland); and EGU General Assembly, Vienna, oral presentation (on the High Börzsöny).

3 Summary of publication activity and conference attendance

As listed under the publications, a great number of scientific communications and conference presentations have appeared during the project run.

A) In total, *18 peer-reviewed papers* emerged containing the obtained results of the research work. Of them, 16 have been published (between 2015 and 2019), and 2 are under review (with expected publication date 2020); 14 of the total are in impact-factored journal. *Cumulative impact factor* of the 18 papers is *27.108*.

Most cited papers (as of 30 December 2019): *Karátson et al. (2016) 20; Veres et al. (2018) 8; Biró et al. (2016) 4; Wulf et al. (2016) 3.*

B) In addition, *1 PhD dissertation, 1 field trip guide chapter* and *1 manuscript report* were also produced.

C) *49 conference presentations* with printed or online available abstract are recorded during the project run. Of them, 15 were given at international meetings/conferences. In addition, there were 2 oral presentations with no abstract.

4 Detailed summary of the obtained results by study area

A. Mátra and Bükk foreland; North Hungarian basins

As mentioned in the Introduction, a main objective of the research was inferring erupted volumes and, if possible, calculating eruptive rates. To achieve this, two crucial constraints are required: (1) precise ages (in the project, by combining paleomagnetic data with K-Ar age data), and (2) detailed knowledge on stratigraphy (of well-defined eruptive units). Out of the target study areas, the volcanic stratigraphical features in North Hungary, comprising the Mátra Mts. and the Bükk foreland study areas as well as adjacent sedimentary basins that host Miocene volcanic successions, are poorly known. It became obvious during the project run that e.g. the Gyulakeszi, Tar, or Felnémet tuff formations (cf. Gyalog & Budai 2004) 1) are sometimes poorly identifiable in the field, 2) consist of a diverse succession of pyroclastic units which are not described or poorly known in the literature, 3) are characterised by radiometric ages which are many times ambiguous or burdened with significant uncertainty (cf. Tar Dacite Tuff: Zelenka et al. 2005). Thus, in the study areas, our efforts focused on gaining information on ages and stratigraphical features based on paleomagnetic rotations and new K-Ar dating, detailed field volcanological descriptions, and geochemical correlations.

The results presented below did not appear in a coherent, single paper, although implications were given in a number of papers and conference presentations. Therefore, here we give a relatively detailed overview of the stratigraphic results and implications, also presenting the applied methods. K-Ar ages obtained are considered preliminary and still need refinement, so are not presented in this report. We are convinced that the new results on the area will allow to better constrain the regional stratigraphy, and to make steps toward volumetric calculations in our new NKFIH-OTKA project.

1) Paleomagnetic correlation of ignimbrites and related pyroclastics

The paleomagnetic results of the Mátra Mts. and the Bükk foreland volcanic area (BFVA herafter) before 2015 were mostly obtained from moderately to strongly welded ignimbrites. In the research project the measurements were extended to non-welded facies as well to improve stratigraphical coverage both in lateral and vertical dimensions. In order to obtain reliable paleomagnetic directions from non-welded facies, we also experimented with samples drilled from pumices supported in loose to moderately cemented matrix. To confirm that the paleomagnetic directions determined from pumice blocks are reliable recorders of the direction of the Earth magnetic field during cooling, we made comparison between matrix (just drillable) and pumice blocks in Láztető quarry (near Noszvaj). As a result of stepwise demagnetization, we obtained statistically identical results from the matrix (represented by several independently oriented samples) and from the pumice blocks (several independently oriented samples from a number of pumice blocks). As we also made Curie-point measurements and the pumice samples were thermally demagnetized, we were also able to conclude that the paleomagnetic signal was stable practically up to the Curie-point of the magnetic mineral, close to 600°C (Márton et al. 2019). This suggests that the ignimbrite was very hot during emplacement, thus able to impart uniform remanent magnetization on the pumice blocks (randomly magnetized before hot emplacement).

The above experiments provided a solid basis for considering the results reliable, obtained from several friable or loosely cemented ignimbrite units with massive pumice block-bearing lapilli tuff facies. The following correlations were made based on paleomagnetic rotations (**Fig. 1,** Biró et al. 2017a, Márton et al. 2017, 2018, 2019):

i) Ignimbrite of the abandoned quarry and road to vine cellars at Harsány show the same ~40° CCW rotations, identical to the one at Tibolddaróc (uppermost level of vine cellars).

ii) The ignimbrites cropping out along Mátrabérc (Western Mátra Mts) show a 0-10° CW rotation, indicating that they were emplaced close in time to the ignimbrites at Sirok, Tar – Fehérkő quarry, Bajúsz valley, and Felnémet quarry.

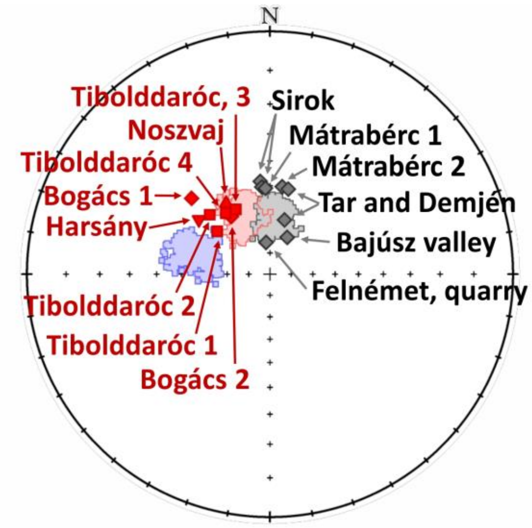


Fig. 1. *New paleomagnetic declinations of ignimbrites and other pyroclastics from the Mátra Mts. and the BFVA (Márton et al. 2019). Coloured zones represent the areas occupied by magnetic directions representing objects which cooled very fast while the direction of the geomagnetic field changed (secular variation during 2000 years from archeomagnetic data: Márton 2010). Grey zone is the present situation, pink and blue zones are those affected by about 30° and 30°+50° CCW rotations, respectively.*

2) Detailed description of a well-preserved phreatoplinian succession

Two occurrences of a BFVA succession (near Bogács and Tibolddaróc, respectively) is inferred to have been produced by silicic phreatomagmatism (phreatoplinian sensu lato) investigated in detail (Biró et al. 2020). Correlation of the two occurrences were based on identical layers and similar, ~30°CCW paleomagnetic rotation and normal polarity (Biró et al. 2017a, Márton et al. 2017, 2018: Tiboldaróc 1, 2, 3, and Bogács 1 and 2 in **Fig. 1**). Such a type of volcanism was documented in detail for the first time in the Carpatho-Pannonian Region. The discontinuously exposed, paleosol-bounded 8.5 m thick phreatomagmatic succession contains three subunits. The first is a 1.8 m thick series of fine to coarse ash layers with laterally constant thickness at outcrop scale and unimodal grain-size distribution. The second is 2.1 m thick and comprises several normal-graded layers with an upper fine-grained zone containing abundant ash aggregates. The uppermost subunit, 4.5 m thick, is a massive, poorly to well-sorted coarse ash with gas escape structures and ash aggregates at its base. The successive change of the recorded lithofacies from base to top implies an initially dry fallout-dominated deposition of ash transforming into a wet one emplaced by multiple pyroclastic density current (PDC) pulses (**Fig. 2**). According to very weak iso-orientation or random distribution of the principal susceptibilities revealed by analyses of low-field anisotropy of magnetic susceptibility (AMS), the energy of the dilute, wet density currents were low (Biró et al. 2017b). The general abundance of PDC-related ash aggregates in the middle-upper part of the succession, and the transformation of a fall-dominated to a collapsing depositional regime producing wet ignimbrites, imply abruptly intervening water influence on a silicic eruption (phreatoplinian sensu lato), similar to e.g. the 26.5 ka Oruanui eruption (Taupo Volcanic Zone, New Zealand; Biró et al. 2020).

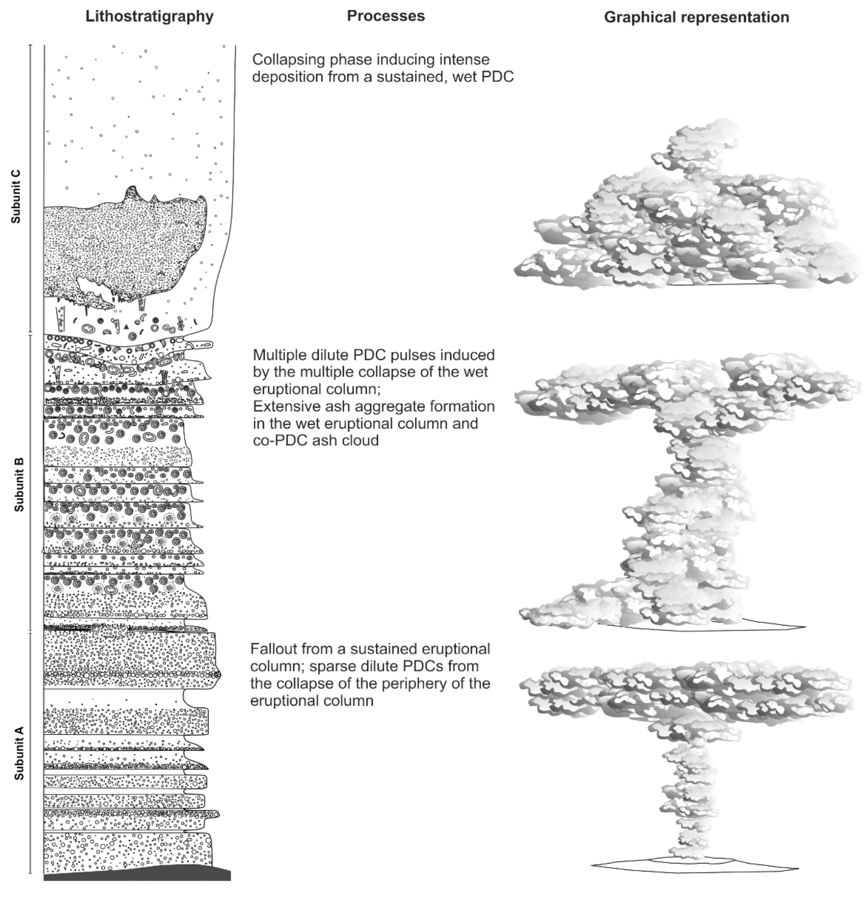


Fig. 2. *Lithological column of the investigated phreatoplinian succession with genetic interpretation (Biró et al. 2020). On the right, graphical representation of eruptional processes which resulted in the formation of various subunits is shown.*

3) A tool for calculating eroded volumes of ignimbrites based on structural hydroxyl content of quartz

It was pointed out that the initial structural hydroxyl content of quartz phenocryst fragments was modified after hot emplacement and slow cooling within two ignimbrite units at the

BFVA (Biró et al. 2017c). The final structural hydroxyl content is a function of cooling speed-moderated diffusional H-loss, which is clearly related to the height above the base of the emplaced ignimbrite. The cooling is instantaneous at the base of the ignimbrites resulting in the preservation of the original structural hydroxyl content (Biró et al. 2016). It is similar at the top of the ignimbrite where cooling is more effective due to contact with the ambient atmosphere, compared to the internal, intact zone. Consequently, with height above the base, the average structural hydroxyl concentration shows a gradual decrease towards the middle of an ignimbrite unit and increases again towards the top. This results in a characteristic structural hydroxyl concentration curve. Such a finding implies that determining the structural hydroxyl content curve vs. height above the base of an ignimbrite (cooling) unit allows to estimate (paleo-)erosional volumes (Biró et al. 2017c, 2019). If the emplacement temperature and cooling rates are known, erosion can be calculated by considering to what extent the curve is removed. In the future, hopefully this method could be used as a unique tool for estimating eroded volumes of ancient ignimbrites.

In addition, based on the above knowledge, structural hydroxyl content of quartz hosted in pyroclastic-fall deposits was investigated, because this pyroclastic deposit type is believed to have cooled much faster than pyroclastics deposited from PDCs resulting in hindered diffusional H-loss. In the BFVA we identified a well-sorted, well-exposed lapilli tuff at three different locations (an eastern, a southern and a western occurrence), which was determined to be the same pyroclastic-fall layer based on physical volcanological features and stratigraphical position. The results show that the structural hydroxyl content of the quartz phenocrysts is identical (Hencz et al. 2019). Hence, the structural hydroxyl content of the quartz phenocrysts could be a potential volcanological correlation tool in case of rapidly cooled pyroclastic deposits. Furthermore, from a regional geological and volcanological point of view, we have found three distinct outcrops in the Bükk Foreland where surely the same part of the whole stratigraphical column crops out, hereby offering an opportunity for future volumetric calculations.

4) Paleontological constraints on the Miocene volcanic stratigraphy in North Hungary

During the project run, efforts were made to collect paleontologically promising material to contstrain the volcanic stratigraphy. Digging of tons of material was conducted in the Visegrád Mountains (Szentendre–Cseresznyés-árok, **Fig. 3**), Börzsöny (Magyarkút), Cserhát (Kisterenye, Gyulakeszi, Nemti, Szentkút–Vadász-gödör, Buják, Kutasó), Mátra (Szurdokpüspöki) and Bükk foreland area (BFVA:

Fig. 3. *Digging during field work at Szentendre–Cseresznyés-árok*

Mikófalva–Őzike-gödör, Bogács, Cserépfalu, Eger – Tihamér quarry).

Only the sites of Cseresznyés-árok, Mikófalva and Szentkút were productive. In Szentendre–Cseresznyés-árok, the positive test resulted in sampling of tons, to be analysed later. At Mikófalva–Őzike-gödör, 100 kg sampled material, two *Albanensia albanensis* teeth were found, proving a Sarmatian age and MN 7+8 vertebrate zone. (In the European Miocene, three species of the flying squirrel *Albanensia* genus were described: *A. sansaniensis, A. albanensis* and *A. grimmi*. *A. sansaniensis* is a characteristic element of the MN5+6 faunas, *A. albanensis* is typical in MN7+8 zone. *Albanensia grimmi* was found in the latest MN7+8 and MN9 zones.) The marine fossils of Szentkút–Vadász-gödör yielded a possibly early Badenian age because of the occurences of the foraminifers *Planostegina costata, Lenticulina inornata, Quinqueloculina* sp. and the Ostracoda *Aurila* cf. *angulata* (in preparation; preliminary findings in Szuromi-Korecz 2018).

The elaboration of the faunas of Kozárd and Vércsorog was completed during the project run, whereas the study of the Cseresznyés-árok fauna is currently going on. Its fauna has direct volcanic stratigraphical impication, since it was collected from a diatomite directly underlying the volcanic succession of the Visegrád Mountains. In the fauna the dominant element is *Cricetodon aureus*. In the Northern Alpine Foreland Basin, this fossil rodent is found in the faunas between the “Brock horizon” and the “Main bentonite horizon”, with a radiometric age between 14.7 -15.0 Ma (Hir & Venczel 2018).

In the well-logs of boreholes in the vicinity of Szentendre, Fót and Budapest, the diatomaceous claymarl is exposed directly under the “Middle Dacite Tuff” (Hir & Venczel 2018).

The freshwater sediments of the Felsőtárkány Basin contain seven vertebrate faunas. This complex is bedded above the “Felnémet Rhyolite Tuff”. Biochronological position is late MN 7+8 zone; radiometric age is estimated between 11.6 -11.1 Ma (Hir et al. 2016, 2017).

In Litke, a lagoon sediment crops out which could be related to the “Tar Dacite Tuff”. Typical element of the vertebrate fauna is *Cricetodon meini*. In the Northern Alpine Foreland Basin (Bavarian and Swiss molasse), this rodent is found in the faunas immediately under the “Brock horizon”. Its estimated radiometric age is 15.2 -15.0 Ma (Hír 2013).

In Hasznos, a diatomite layer is embedded in the “Nagyhársas Andesite”. The study of the locality was initiated by László Kordos during the 1980’s. We collected additional finds including the study of the Insectivor and Chiropteran material (Hír et al. 2016, 2017, Rosina et al. 2015, Prieto et al. 2015). The most characteristic element of the fauna is *Cricetodon hungaricus* with a biochronological position MN6. The systematic revision of the *Cricetodon* material was presented in Hír (2017).

In Sámsonháza, a lagoon sediment crops out above the marine “Sámsonháza Formation” and under the “Nagyhársas Andesite”. The mollusc-rich material was studied by the late József Kókay who classified the fauna as middle Badenian, showing a vertebrate assemblage MN6. The biochronological position of the vertebrate faunas of Hasznos and Sámsonháza should be very close to each other (Hír et al. 2016, 2017).

The faunas of Kozárd and Vércsorog were excavated and elaborated during the project run. From the biochronological point of view, these faunas are highly important because fossils of marine species and fossils of dry land vertebrates are equally found (Hír et al. 2019). Moreover, these localities imply a unique opportunity for the harmonization of the marine and continental stratigraphy. The faunas verified the correlation of the marine “*Elphidium reginum* zone” and the continental MN7+8 Zone (Hír et al. 2019).

Obviously, the above findings should significantly contribute to refining the North Hungarian volcanic stratigraphy in integrated papers, this being one of the main objectives of the new NKFIH-OTKA project.

B. Börzsöny Mts.

In the Börzsöny–Visegrád Mts. study area, we focussed on the Mid-Miocene volcanism of the Börzsöny, in particular to the time-space evolution of its largest edifice, the High Börzsöny (Karátson et al. 2000). The timing of the volcanism has been re-evaluated by correlating new New Cassignol-Gillot K-Ar dating with paleomagnetism (Karátson et al. 2018a, 2019a, b). A manuscript with the new age data (i.e. volcano growth) and constraints on magma output rates, based on conference presentations given above (see publication list), is being formulated.

The volcanism of the Börzsöny Mts. developed in three stages. The first-stage activity occurred in a shallow marine environment, producing mostly dacitic volcaniclastics (originating from small-scale ignimbrite eruptions and lava domes). In our study, we re-measured the age of the Királyrét Ignimbrite by Gillot-Cassignol K-Ar dating (**Fig. 4**). The age was obtained on biotite from dacitic pumiceous lapilli tuff and yielded 15.68±0.22 Ma (1σ), which largely fits to the magnetostratigraphically established age (≥16 Ma) of the first stage (normal polarity, 30^{o} CCW rotation). The second stage, not studied, was characterised by scattered andesitic lava domes. Importantly, during this stage, a ca. 30^{o} CCW rotation occurred

Sample no.	Locality, dated fraction	*Weighted mean age (Ma, ±1σ)*
16BOR47	Ökör-orom andesite lava, groundmass	*14.27±0.20*
16BORIQ39	Inóc Quarry andesite lava, groundmass	*14.79±0.21*
19BORNH	Nagy-Hideg-Hill, andesite lava flow, groundmass	*14.87±0.21*
19BORSZB	Száraz-bérc andesite lava, groundmass	*14.98±0.21*
17BOR01	Csarna-patak W andesite lava, groundmass	*14.99±0.23*
19BORTV	Templom-völgy andesite lava, groundmass	*15.01±0.21*
15BORK1	Királyrét, ignimbrite dacite lithoclast, biotite	*15.68±0.22*

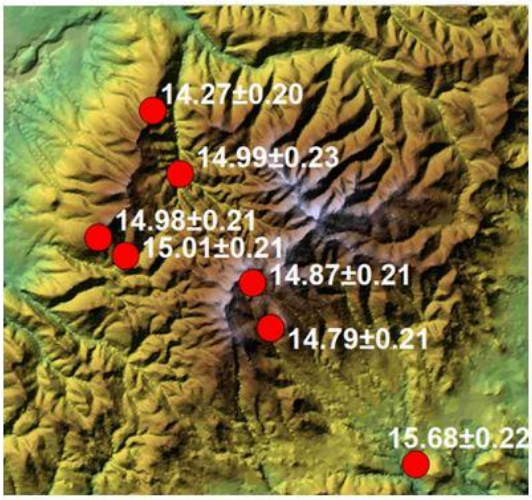


Fig. 4. *Unpublished, preliminary K-Ar ages of the Mid-Miocene volcanism of the Börzsöny Mts. without analytical data (table and coloured DEM with sampling sites). Note that the majority of the ages were obtained on groundmass of the High Börzsöny andesite lava rocks, whereas the age of the Királyrét dacitic ignimbrite (representing the first volcanic stage) on biotite separate.*

During the third stage, a moderately explosive, andesitic / basaltic andesitic subaerial lava dome complex, called High Börzsöny, was produced, consisting of lava flows and block-and-ash flows which show no rotation. The regular-shaped dome complex has developed a central, enlarged erosion caldera, clearly recognisable in the landscape despite long-term denudation and intense tectonic faulting over the mountains. The new Cassignol-Gillot K-Ar dating mainly focussed on this stage, namely the groundmass of the andesitic lava rocks of the High Börzsöny. The obtained ages, ranging between 15.01±0.21 and 14.27±0.20 Ma, are in agreement with their stratigraphic position from bottom (the internal part of the High Börzsöny erosion caldera) to top (flank lava flows), and are somewhat (ca. 0.5 Ma) older than previous conventional K-Ar measurements (Karátson et al. 2000).

Combining the newly obtained ages with paleomagnetism constrains the activity of the High Börzsöny, which, subsequent to a reverse polarity zone (of the second stage), was active within one or two normal polarity zones ending >14 Ma. The time interval for the High Börzsöny dome complex (0.74±0.31 My) fits to other well-known domes worldwide (e.g. Montagne Pelée, 550 ky).

Whereas uncertainties, to be minimized in subsequent analyses, allow a shorter life time as well (<0.5 My), a longer duration is not likely. The new ages will make it possible to calculate eruptive rates during the life time of the volcano.

C. East Carpathians

The East Carpathian volcanic range (**Fig. 5.**) experienced an along-arc, Late Miocene to Quaternary migration of eruptive activity. During the project run, after an extended field sampling campaign for K-Ar dating, a novel and complex methodology was applied to produce new geochronological and geomorphological constraints on the volcanic and erosional dynamism. Apart from results on individual volcanic edifices, magma output rates (km^3/My) as well as erosion rates (m/My) for the whole volcanic range were calculated. The obtained results were submitted by Dibacto et al. 2020, to Geomorphology (under review), and summarized in some details as follows.

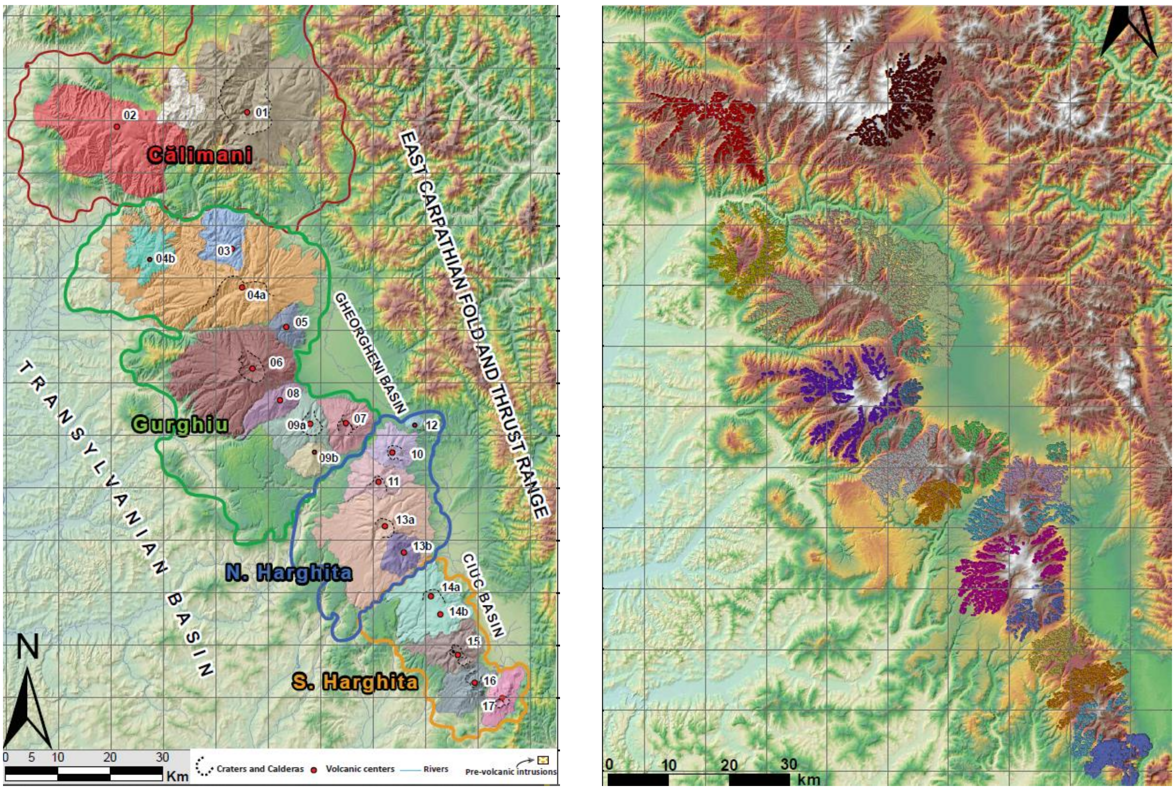
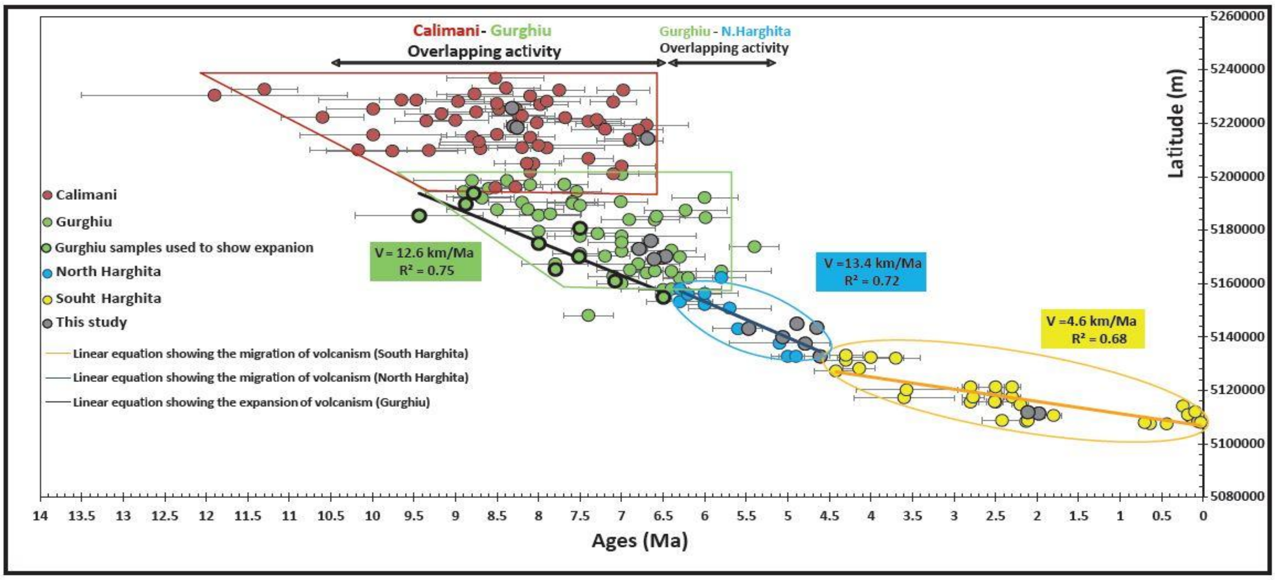


Fig. 5. *Geographical setting of the East Carpathian volcanoes (left) with constraining points of their surface used for morphometric reconstruction (coloured dots; right)(Dibacto et al. 2020)*

For four volcanoes (Central Călimani / Központi Kelemen, Seaca-Tătarca/Mezőhavas, Vârghiş/Madarasi-Hargita, Pilişca/Piliske edifice), new ages were obtained applying the unspiked Cassignol-Gillot K-Ar technique to determine either their lifespan (6.79±0.10–6.47±0.09 Ma for Seaca-Tătarca; 5.47±0.08–4.61±0.07 Ma for Vârghiş); the end of volcanic activity (e.g. a new, youngest age of 6.69±0.10 Ma for Călimani); or refine previous conventional K-Ar ages.



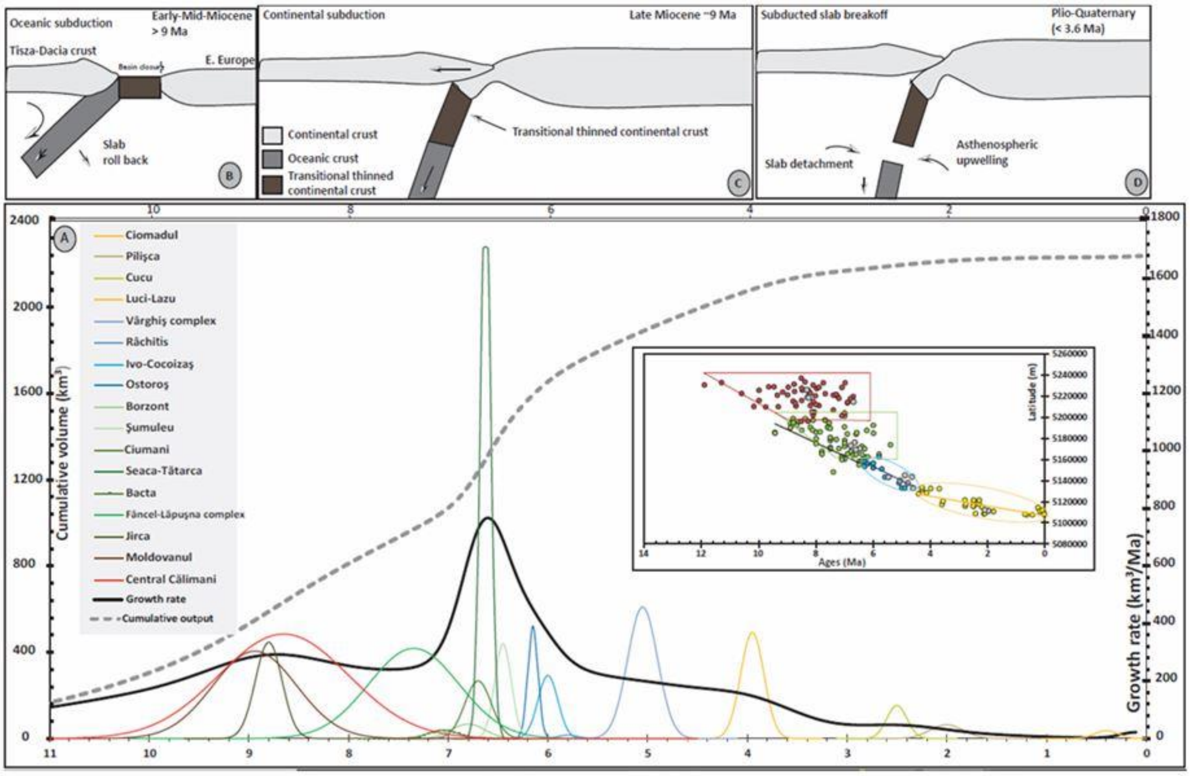


Fig. 6. *Upper figure: age progression along the East Carpathian volcanic range (quadrangles show km/Ma values for the Gurghiu/Görgényi, North and South Harghita/ Hargita segments); lower figure: cumulative volume through time (main graph, A) correlated with main Tertiary tectonic events affecting the East Carpathians (displayed as cartoons B, C and D), and individual variations (at volcano scale) of magma output through time (km³/My) defined by Gaussian-like curves (coloured lines; Dibacto et al. 2020)*

In parallel, numerical reconstruction of volcanic paleo-topographies was performed by using an improved algorithm (ShapeVolc, developed in GEOPS, Orsay) which allowed to model their shape prior to any major dismantling processes, i.e. reconstructing the initial topography that they reached at the end of edifice growth (called pre-erosional volcanic surface). On this basis, individual volumes of the twenty identifiable East Carpathian volcanoes were quantified, showing a wide range of volcano size ranging from 3±3 to 592±115 km^3.

A total constructed volume of ~2300 km^3 making up the four main segments of the East Carpathian volcanic range (910 km^3 for Călimani, 880 km^3 for Gurghiu, 279 km^3 for North Harghita and 165 km^3 for South Harghita segment) was computed. New volumes, coupled with available ages, allow to compute an average growth rate of 200 km^3/Ma for the entire range, characterized in detail by two main trends: an initial moderate growth rate (137 km^3/Ma) for the northern, older, Mio/Pliocene volcanoes (Călimani-Gurghiu-North Harghita and Luci-Lazu) followed by a drastically lower growth rate (28 km^3/Ma) obtained for the mostly Plio/Quaternary volcanoes of the South Harghita segment. Out of these trends, we computed the highest value of growth rate for Seaca-Tătarca volcano (825 km^3/Ma). This finding may be however due to the higher precision of the new unspiked K-Ar ages, which makes it likely that other volcanoes also grew up faster than constrained by previous age data.

A total eroded volume of 524±125 km^3 was computed for the entire range, resulting in an average degree of denudation of 22% and an average erosion rate of 15 m/Ma (17 m/Ma for Călimani; 11 m/Ma for Gurghiu; 9 m/Ma for North Harghita; and 20 m/Ma for South Harghita), evidencing a variable intensity of erosion through time. Specific erosion rates for each major climatic period have been computed, which fit well with the corresponding climatic context: the highest erosion rate (38 m/Ma) occurred during the first, transitional moderate subtropical-continental climate period (9.5 and 8.2 Ma); an intermediate erosion rate (13 m/Ma) characterized a moderate continental climate period up to 6.8 Ma when conditions became less humid; an extremely low erosion rate (6 m/Ma) was pointed out for a prevailing continental but occasionally semi-arid climate (6.8-5.8 Ma); and finally, again a higher erosion rate (22 m/Ma) was derived for the Plio/Quaternary testifying the erosional impact of rapid shifts between wetter and colder conditions in the interglacial/glacial cycles that generally characterize this period.

Our quantitative morphometric and geochronological approach demonstrates that reconstructing volcanic paleo-topographies is an efficient tool to study the volcanic dynamism through time, also yielding implications to the tectonic and climatic contexts that drive both constructional and dismantling processes at arc-type volcanic settings such as the East Carpathian range.

D. Ciomadul (Csomád)

During the project, special attention was devoted to the complex study of Ciomadul volcano, the youngest (Late Quaternary) edifice of the whole Carpatho-Pannonian area. This was in part motivated by the internationally growing interest toward this peculiar volcano, attracting several working groups. In our approach, we focussed on the long-term time-space evolution of the lava dome complex in the light of magma output rates (published in most detailed form by Lahitte et al. 2019, Karátson et al. 2019c), and the most exciting late-stage evolution up to <30 ka, characterized by a regionally significant explosive activity (Karátson et al. 2016, 2017, Wulf et al. 2016, Veres et al. 2018). Since the obtained results have been published in full, here we give only a general overview of the main findings.

1) Long-term time-space evolution

The volcanic field of the dacitic Ciomadul developed on the erosional surface of Lower Cretaceous flysch and ~2 Ma old andesites and experienced an extended eruptive history from ~850 to <30 ka. New eruption ages were determined following a strict separation (of 10 g) of groundmass from about 3 kg of unaltered lava rocks, thereby isolating material whose cooling was contemporaneous with the eruption. The newly applied methodology, mainly consisting of a double full preparation, first at larger grain size (~0.4 mm) and then at <100 μm, provides an appropriate procedure to separate suitable material to obtain the K-Ar age of the eruption, i.e. the sample's groundmass, in which there is no risk of the presence of older, inherited crystals (Lahitte et al. 2019).

Predominantly effusive activity took place during the first stage (~850 to ~440 ka), producing volumetrically minor, isolated, peripheral domes. Subsequently, after a ~50 ky repose interval, a voluminous central dome cluster developed in the second stage (~200 to <30 ka). During the youngest phase of evolution (~60 to <30 ka), highly explosive eruptions also occurred, resulting in the formation of two craters (Mohos and St. Ana). Combining K-Ar ages with an advanced DEM-based morphometric approach resulted in the following findings (Karátson et al. 2018b, 2019c). The calculated total volume of the lava domes is ~8.00 ± 0.55 km^3, which includes the related volcaniclastic (1.57 km^3) as well as erosionally removed (0.18 km^3) material, and is in line with dimensions of other medium-sized dacitic lava domes worldwide. This volume was extruded at an average long-term magma output rate of 9.76 km^3/My (0.0098 km^3/ky). However, most of the domes (7.53 ± 0.51 km3) were formed in the 200 to <30 ka period, implying a significantly increased magma output rate of 37.40 km^3/My (0.0374 km^3/ky), more than 30 times higher than in the first stage (**Fig. 7**).

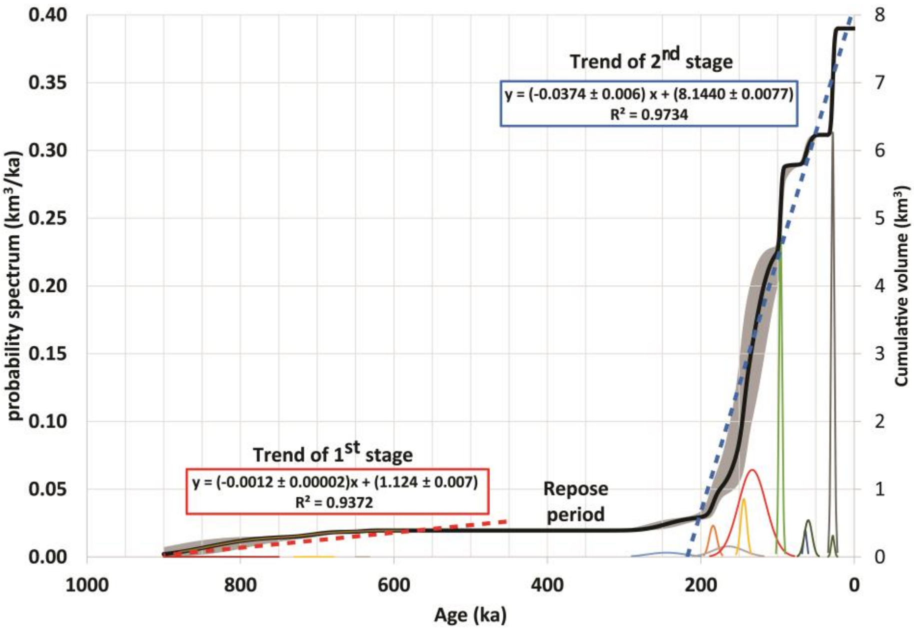


Fig. 7. *Development of the Ciomadul volcanism as reflected by magma output in time (km^3/ka) showing first a moderate and subsequently an increased trend in terms of volume (Karátson et al. 2019c).*

Within these long-term trends, individual lava domes of Ciomadul (e.g. those with volumes between 0.02 and 0.40 km^3) would have been emplaced at much higher rates over a period of years to tens of years. The active periods, lasting up to hundreds of years, would have been followed by repose periods ~30 times longer. The most recent eruption of Ciomadul has been dated at 27.7±1.4 ka, which denotes the youngest age ever measured at the volcano. This age, which is in agreement with radiocarbon dates for the onset of lake sediment accumulation in St. Ana crater, dates fragmented lava blocks which are possibly related to a disrupted dome. This suggests that during the last, typically explosive, phase of Ciomadul, lava dome extrusion was still ongoing (see point 2). In a global context, the analysis of the volumetric dynamism of Ciomadul's activity gives insights into the temporal variations in magma output; at lava domes, short-term (day or week-scale) eruption rates smooth out in long-term (millenia-scale) output rates which are tens of times lower.

2) The explosive late-stage activity

The most recent, mainly explosive eruptions of Ciomadul have been constrained by detailed field volcanological studies, major element pumice glass geochemistry (**Fig. 8**), luminescence and radiocarbon dating, and a critical evaluation of available geochronological data (Karátson et al. 2016, Wulf et al. 2016, Veres et al. 2018). Our investigations were complemented by the first tephro-stratigraphic studies of the lacustrine infill of Ciomadul's twin craters (St. Ana and Mohoş) that received tephra deposition during the last eruptions of the volcano. The approach for a detailed tephrostratigraphy introduced a three-letter abbreviation of field outcrops (e.g. BTS: Baile Tusnad), highly useful for identification, and generally adopted in subsequent literature.

Our analysis showed that a significant explosive activity, collectively called EPPA (Early Phreatomagmatic and Plinian Activity), started at Ciomadul in or around the present-day Mohoş, the older crater, at ≥51 ka BP (Karátson et al. 2016). Later K-Ar dating (Lahitte et al. 2019) made it likely that the first explosive crater-forming eruptions may have been older (>130 ka?). These explosive eruptions resulted in a thick succession of phreatomagmatic fall deposits (e.g. Turia Tuffs) found in both proximal and medial/distal localities around the volcano, characterized by highly silicic (rhyolitic) glass chemical compositions (ca.75.2–79.8 wt.% SiO_2). The EPPA stage was terminated by a subplinian/plinian eruption around 43 ka BP, producing pumiceous pyroclastic-fall and -flow deposits of similar glass composition, probably from a "Proto-St. Ana" vent located at or around the younger crater (hosting the present-day Lake St. Ana).

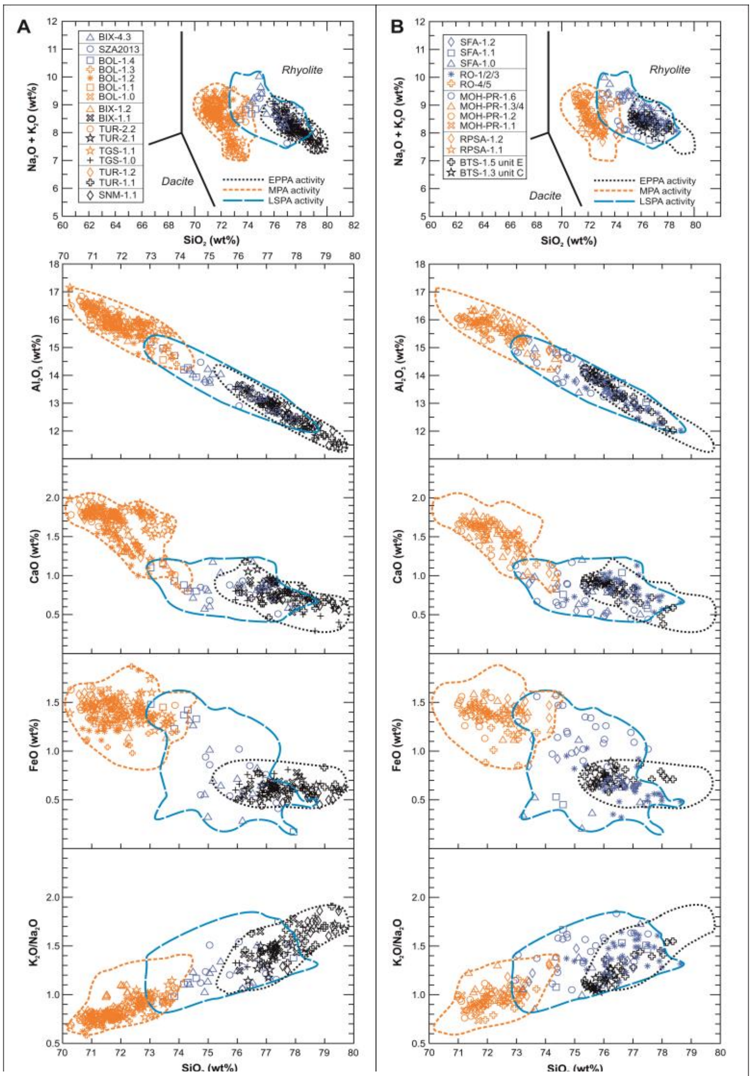


Fig. 8. *Discrimination of three main phases (EPPA, MPA, LSPA) of Ciomadul's last explosive stage as defined by bi-variate plots of major element glass compositions of juvenile components (Karátson et al. 2016, Wulf et al. 2016)*

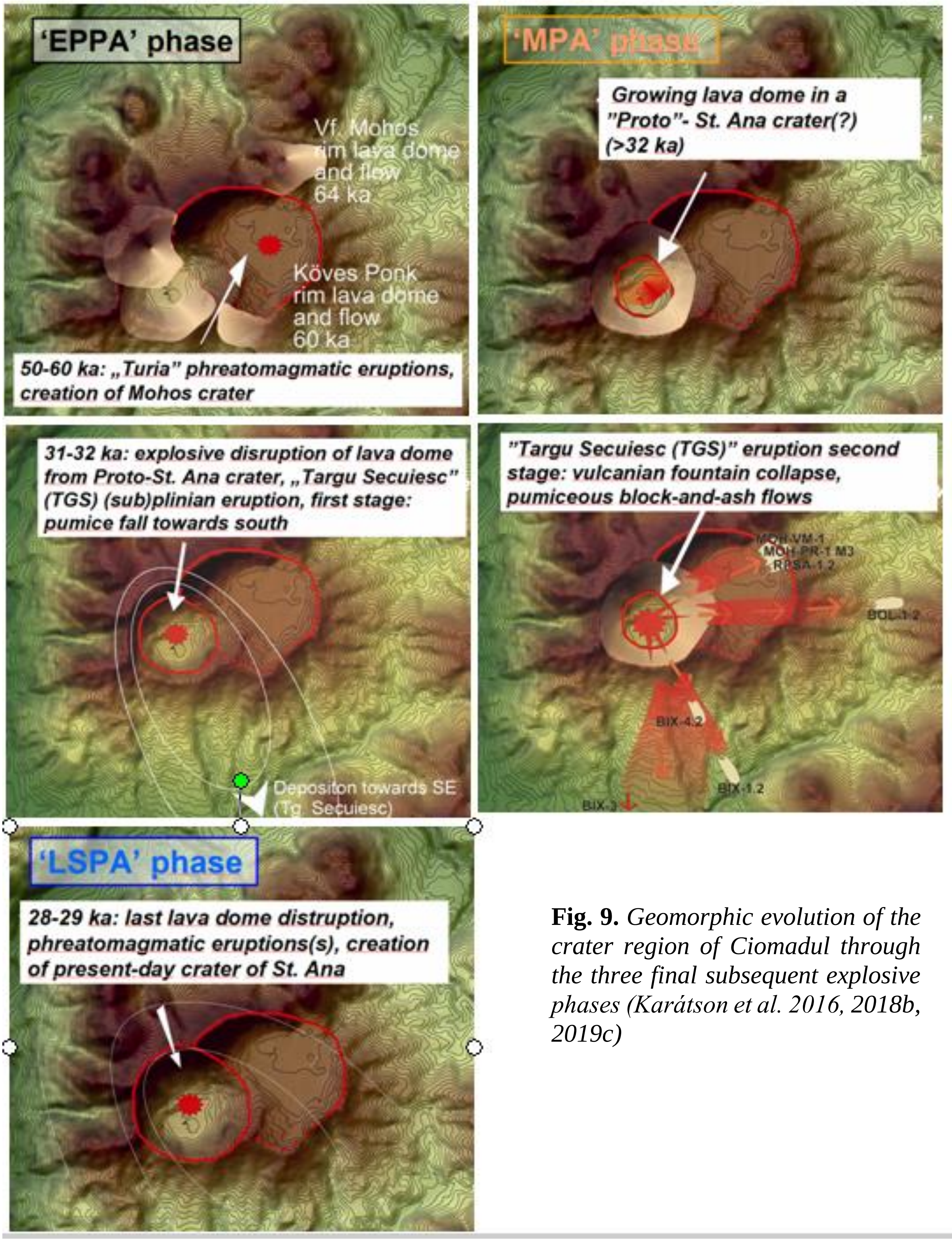


Fig. 9. *Geomorphic evolution of the crater region of Ciomadul through the three final subsequent explosive phases (Karátson et al. 2016, 2018b, 2019c)*

After a quiescent period with a proposed lava dome growth in the St. Ana crater, a new explosive stage began, defined as MPA (Middle Plinian Activity: Karátson et al. 2016). In particular, a significant two-phase eruption occurred at ~31.5 ka BP, producing pyroclastic falls (possibly disrupting the preexisting lava dome) and then pyroclastic flows from vulcanian explosions. Pyroclastic deposits of MPA show a characteristic, less evolved rhyolitic glass composition (ca. 70.2–74.5 wt.% SiO_2) and occur both in proximal and medial/distal localities up to 21 km from source.

The MPA eruptions, that may have pre-shaped a crater similar to, but possibly smaller than, the present-day St. Ana crater, were followed by a so far unknown, but likewise violent last eruptive phase from the same vent, creating the final morphology of the crater. This phase, referred to as LSPA (Latest St. Ana Phreatomagmatic Activity), produced pyroclastic-fall deposits of more evolved rhyolitic glass composition (ca. 72.8–78.8 wt.% SiO_2) compared to that of the previous MPA stage. As mentioned above, more recent K-Ar dating on brecciated lava blocks close to St. Ana crater yielded a 27.7±1.4 ka age, directly confirming the age of the latest explosive activity at Ciomadul (i.e. subsequent to MPA: Karátson et al. 2019c). Radiocarbon age constraints on bulk sediment, charcoal and organic matter from lacustrine sediments recovered from both craters, as well as radiocarbon ages obtained from St. Ana crater (ca. 27 ka), are in agreement with the timing of this youngest phase.

The above findings made it possible to infer the development of crater morphology in the central part of the volcano. Subsequent growth of lava domes, their explosive disruption, truncation of former lava domes and formation/enlargement of craters (first, the larger Mohos, then the smaller but deeper St. Ana), all of which can be connected of the EPPA, MPA and LSPA phases as defined by tephrostratigraphy, were identified as the most important geomorphic changes. Snapshots of selected, subsequent events as shown on DEM images are depicted in **Fig. 9.** (Karátson et al. 2016. 2018b, 2019c).

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