Final research report

NKFIH/OTKA PD 112584 project

Origin and evolution of the Miocene silicic magmatism in the Pannonian basin primarily based on textural and compositional characteristics of zircon from the Bükkalja ignimbrites

2014 - 2018

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Introduction

Eruptions of silicic (SiO_2 > 65 wt%) magmas are among the most devastating volcanic events in the Earth and in the last decade, the nature and reason of such volcanism are in the frontline of research. One of the key-issues in these studies is the origin of the silicic magmas and the emplacement and longevity of the silicic magma reservoirs in the continental crust. In the Pannonian basin, large volume of silicic magmas erupted during the Early to Mid-Miocene that resulted in widespread volcanic material, which has been used as stratigraphic key-horizons for decades. Traditionally, these silicic volcanic products have been divided into three main horizons (Lower rhyolite tuff at 21.0-18.5 Ma; Middle dacite tuff at 17.5-16.0 Ma and Upper rhyolite tuff at 14.5-13.5 Ma; Ravasz, 1987, Hámor et al., 1980 and Márton and Pécskay, 1998), although there have been growing evidences that these cannot represent single eruption events and also their accurate ages have been unresolved. This volcanic activity occurred during the formation of the Pannonian basin, when the lithosphere thinned dramatically. Such tectonic setting, i.e. large volume silicic volcanism in an extensional setting is relatively rare worldwide. However, the relationship between the Early to Mid-Miocene magmatic and tectonic events in the Pannonian basin have not been studied in detail.

In the research proposal submitted in the beginning of 2014, we addressed a number of key-questions related to the Early to Mid-Miocene silicic volcanism of the Pannonian basin and intended to answer these using primarily a zircon-perspective study. The advantage of the zircon-based research is that we could obtain accurate dates for the eruption chronology as well as for the longevity of the magma storage in the crust and also on the nature of the silicic magmas, i.e. the role of mantle and crustal components. One of the difficulties of the study of the silicic volcanism in the Pannonian basin is that most of the volcanic products are covered by post-volcanic sediments due to the subsidence of the region. However, the Bükkalja volcanic field (BVF) provides an excellent natural laboratory for studies of the silicic volcanic rocks since the outcrops cover the entire volcanic period and the volcanic formations are mostly fresh. Thus, our study focused primarily on the main volcanic units exposed in this area that could represent the proximal deposits. We sampled further outcrops near the Bükkalja area and some other key-localities, such as Ipolytarnóc and the surroundings and the Mecsek Mts. in southern Hungary. In addition, we collected samples from drilling cores from boreholes in the northern Great Hungarian Plain that could enable to correlate the proximal deposits as well as a couple of samples from the Tokaj Mts. for comparison.

Samples analysed and methods used during the study

During our NKFIH-supported research project, we analysed more than 1500 zircon crystals from more than 30 samples, which thought to cover the entire Early- to Mid-Miocene silicic volcanism in time and space. Most of the samples represent the volcanic formations of the Bükkalja volcanic field (Figure 1.; Lukács et al., 2018).

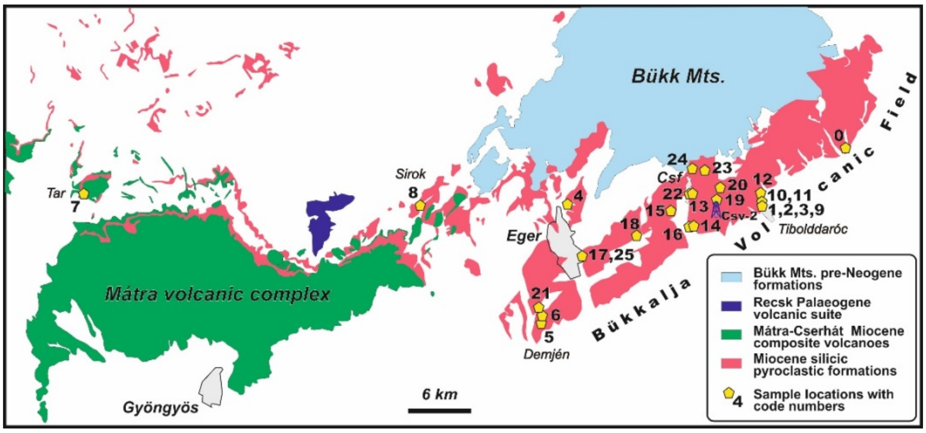

Figure 1. Locations of the studied samples in the Bükkalja volcanic field and in the surrounding area

We conducted geochronological study using LA-ICP-MS U-Pb isotope dating technique in the ETH Zürich, Switzerland in collaboration with Professor Olivier Bachmann and his research group and in the Göttingen University in collaboration with István Dunkl. Each isotope data was carefully evaluated and was used to calculate the U-Pb date and the uncertainty. We collected 30-50 spot analyses from zircons of each samples. The spread of the U-Pb dates and their uncertainties were used to estimate the longevity of the magma storage, whereas we used the youngest U-Pb date population to infer the eruption age. In order to validate this method and obtain an accurate eruption age, we performed high-precision CA-ID-TIMS and Ar-Ar geochronological analysis on the representative samples of the selected main volcanic units. In situ Hf isotope analysis on a subset of samples was carried out by MC-IPC-MS at ETH Zürich. (U-Th)/He dating was performed in collaboration with Martin Danisik (Australia). Chemical composition of pumices and bulk rocks was determined at the ACME Labs, Canada. At the end of the research project, we determined the Oxygen isotope composition of the zircons and coexisting mineral phases at the University of Oregon in collaboration with Professor Ilya Bindeman. Mineral and glass compositions from different units of the silicic volcanic rocks found in Bükkalja were analysed by microprobe at the Göttingen University. The structural behaviour of zircons was checked by Raman spectroscopy at the Eötvös University.

Brief summary of the main results

We published most of our results in four peer-reviewed journals and presented them in several conferences. Additionally, the obtained results were involved also into further publications. Within them, the most significant papers were published in the Contributions to Mineralogy and Petrology (D1 journal with IF: 3.626; Lukács et al., 2015) and in the Earth-Science Reviews (D1 journal with IF: 7.491) as an Invited Review paper (Lukács et al., 2018a). All geochronology data of the Bükkalja zircons were published in the Data in Brief Open Access journal (Lukács et al., 2018b). In the following, we briefly summarize only the main results of our research.

1. We conducted the first high-precision zircon U-Pb dating on the Miocene silicic volcanic rocks of the Pannonian basin. Interpretation of the LA-ICP-MS U-Pb dates was validated by accurate CA-ID-TIMS ages. Furthermore, we performed also (U-Th)/He zircon dating, the obtained data gave the eruption ages, suggesting that no subsequent heating or significant subsidence affected the studied silicic volcanic products.
2. All the data about zircons from the Bükkalja are published in the open access journal of Data in Brief and thus, it provides a data base for further studies such as correlation with distal areas and provenance research (e.g., Kelemen et al. 2017), among others.
3. The length of the volcanism was shorter than previously thought: it was less than 4 Ma from 18.2 Ma to 14.4 Ma based on the Bükkalja and other studied samples.
4. We could distinguish 8 main eruption phases and within that, 3 eruptions could have been large enough to cover extended area by volcanic ash. These volcanic deposits have a key significance in regional stratigraphy: (1) Harsány ignimbrite at 14.358±0.015 Ma; (2) Demjén ignimbrite at 14.880±0.014 Ma; and (3) Mangó ignimbrite at 17.055±0.024 Ma. Thus, the former rigid triple division of the pyroclastic succession has to be replaced with these new results. The former Lower rhyolite tuff (Gyulakeszi Rhyolite Tuff) could correspond to the Mangó ignimbrite unit, the Middle dacite tuff (Tar Dacite Tuff) could correspond to the Demjén ignimbrite unit, whereas the Harsány ignimbrite unit could correspond to the Upper rhyolite tuff (Galgavölgy Rhyolite Tuff). In addition, eruption resulted in the Bogács unit at 16.816±0.059 Ma was also significant forming pyroclastic flows with welded facies overlain by a subunit with strongly heterogeneous juvenile clast population, a feature what was detected also in some boreholes south of Bükkalja.
5. Our determined eruption ages shed new light on the major block rotation events defined by palaeomagnetic rotation data (Márton and Pécskay, 1998; Márton et al., 2007). Following the former interpretation of the rotations by Márton and Pécskay (1998), i.e. they occurred between large volcanic events, we refined the age of them: the first one should have occurred between 17.055 and 16.816 Ma (50 degree CCW), whereas the second one between 16.2 and 14.88 Ma (30 degree CCW).

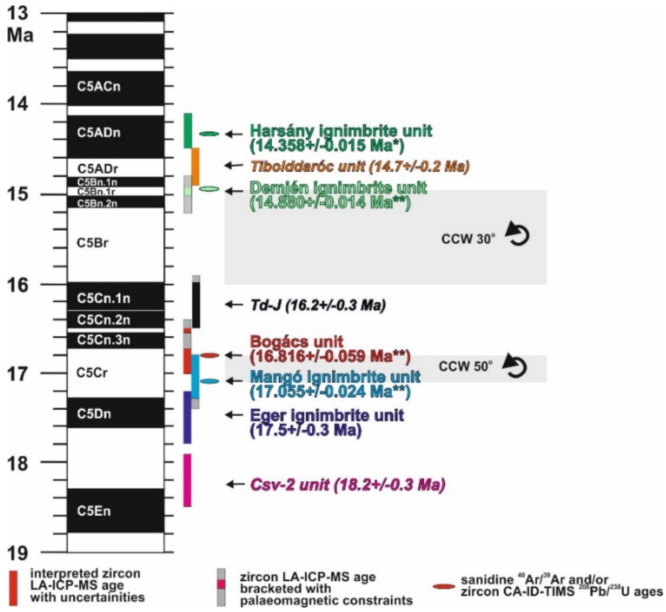

Figure 2 – New subdivision of the eruption phases within the Early to Mid-Miocene silicic volcanism of the Bükkalja volcanic field based on the new high-precision U-Pb zircon dates

6. One of the largest eruptions could have yielded the Demjén ignimbrite unit of the BVF: it involves the ignimbrites found at Tar (Fehérkő quarry, NW Mátra), at Sirok (NE Mátra), at Demjén and also at Tibolddaróc. In addition to the concordant eruption ages, this correlation is supported by the similar normalized trace element distribution patterns and zircon chemistry.
7. The youngest large eruption event was the Harsány ignimbrite occurring in the eastern part of Bükkalja. It has a distinctive geochemical, including zircon trace element fingerprint compared to the Demjén ignimbrite, enabling the differentiation of their volcanic products.
8. Three eruptions (Harsány ignimbrite, Demjén ignimbrite and Mangó ignimbrite) could have expelled several hundreds of km^3 volcanic material, the volcanic ash could spread more than 1000 km distance and accumulated in the sedimentary successions of Paratethys. Thus, they can be used as important marker layers in the Paratethys chronostratigraphy. Further zircon perspective correlation studies could refine this important regional chronostratigraphic framework.

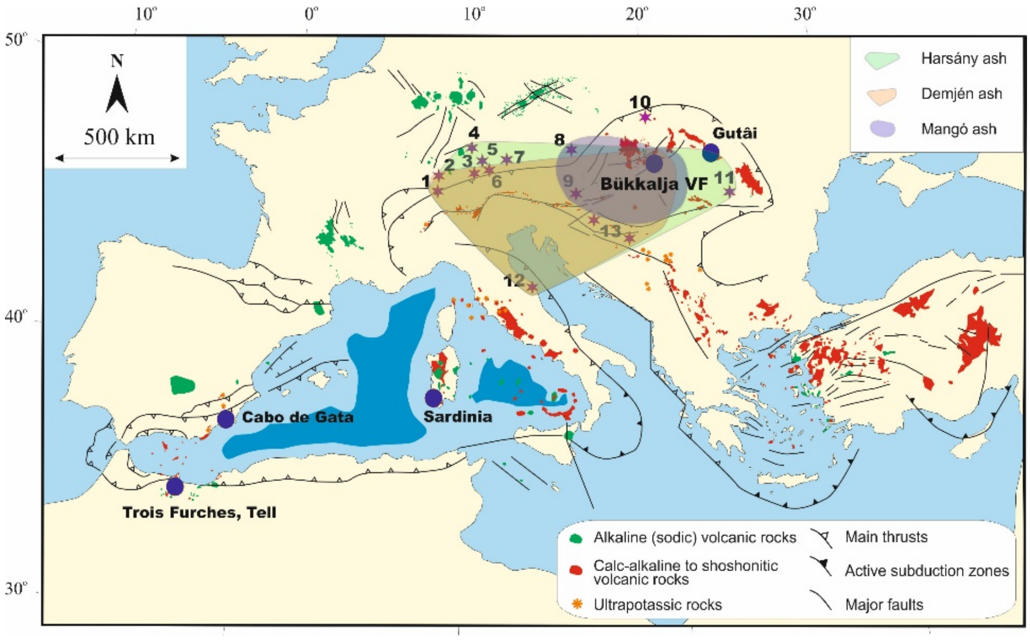

Figure 3 – Extent of volcanic ash deposits of the three main eruption units in central and southern Europe based on the new high-precision U-Pb zircon dates

9. The cumulative volume of the volcanic material formed during this 4 Myr long silicic volcanism is estimated to be>4000 km^3, consistent with a significant ignimbrite flare-up event. These eruptions are regarded one of the most volcanic events in Europe for the last 20 Myr.
10. Mafic magmas are considered to have been formed by melting of the thinned lithospheric mantle metasomatized previously by subduction-related fluids and emplaced at the crust-mantle boundary. They evolved further by assimilation and fractional crystallization (AFC) to generate silicic magmas, which ascended into the pre-warmed upper crust and formed extended magma storage regions.
11. The silicic magma systems fed the main eruption phases could have a prolonged – several 100s thousands of ka - existence in the crust. Magma storage with distinct magmas could

have emplaced simultaneously in the upper crust. Overall, the continental crust beneath the Pannonian basin was thermally strongly affected during the Early to Mid-Miocene.

12. The silicic volcanism in the Pannonian basin was coeval with thinning of the lithosphere and the continental crust. Zircon Hf isotope and bulk rock Sr-Nd isotopic data indicate a sharp decrease of crustal contribution and/or increase of asthenospheric mantle input after 16.2 Ma, suggesting that by this time the crust, and the lithospheric mantle was considerably thinned.
13. The silicic magmatism appears to have had a structural relationship to tectonic movements characterized by strike-slip and normal faults within the Mid-Hungarian Shear Zone (Petrik et al., 2016; Lukács et al., 2018), as well as vertical axis block rotations, when the two microplates were juxtaposed. This volcanism shows many similarities with other rift-related silicic volcanic activities such as the Taupo Volcanic Zone (New Zealand) and the Basin and Range Province (USA).
14. The Early to Mid-Miocene massive silicic volcanism resulted in volcanologically well-preserved formations, which are important geoheritage of our country and thus, they could play important role in geotourism and promotes geopark initiatives as well as could be part of the proposed Pannonian Volcano Route (Szepesi et al., 2017; 2018).

Furthermore, there are some additional, yet partly unpublished important results related to this research project, which are the base of two under preparation publications:

1. The volcanic deposits found in Ipolytarnóc, Nemti, Mátraszele as well as in the northern Mecsek (Máza) could be correlated to the Mangó ignimbrite unit. Volcanic deposits in the Mecsek (Hetvehely, Hidas, Mecsekjános) could belong to the Demjén ignimbrite unit. We assume correlation between the Harsány ignimbrite and the silicic volcanic deposits of the Dés tuff unit based on the same eruption ages.
2. A pre-warming stage, what is necessary for the development of large silicic magma bodies in the upper crust, could be identified before the onset of the silicic volcanism: this was characterized by formation of an extended lava dome field along the Mid-Hungarian Shear Zone. We determined the ages of the volcanic rocks drilled around Paks and obtained 18-20 Ma. The andesitic-dacitic volcanic rocks of the area show strong geochemical similarities with the Early Miocene andesites found in the Mecsek Mts. This volcanism could be related to the onset of the syn-rift phase of the Pannonian basin.
3. The silicic volcanism occurred later in the Tokaj Mts. as our preliminary zircon U-Pb ages imply and further zircon perspective research could help to refine this volcanic activity what resulted in caldera-forming eruption events as well as lava dome building stages (Szepesi et al. 2018, submitted).
4. Our zircon perspective research experiences helped in other on-going studies such as constraining the magma chamber processes beneath Ciomadul, the youngest volcano of the Carpathian-Pannonian region. Furthermore, we used our geochronological experiences to determine the eruption ages of Permian silicic volcanic rocks in southern and western Hungary (Szemerédi et al. 2017 and 2018, submitted).
5. Major and trace element composition of glasses as well as mineral phases appear to be diagnostic of distinct volcanic units and therefore can be used as correlation tools in proximal areas. The zircon trace element signatures are also characteristic of the eruption units and correlate with glass trace element data. Therefore, zircons could have a primary importance in correlation with distal deposits in the lack of fresh juvenile mineral and glass phases.

Acknowledgements

The results of this research could not be achieved without the financial support of the NKFIH/OTKA. It enabled to create strong scientific collaboration with leading scientists and to make our volcanological, geochronological and petrological work more visible in both national and international level. It helped to point out that the volcanic formations in Hungary have a strong scientific as well as geoheritage values. Their studies could help to get a better knowledge on the nature of large silicic volcanic eruptions and their pre-eruption magma storage processes. Our results get strong national and international interest and already initiated collaborative studies with scientists from central, southern and eastern Europe.

Presentation of the results during the project years

Oral presentations:

In Hungarian scientific meetings or conferences:

2015: **2** (MFT Általános Földtani Szakosztály ülés, VI. Kőzettani és Geokémiai Vándorgyűlés); 2016: **2** (VII. Kőzettani és Geokémiai Vándorgyűlés, MTA Tudományos ülés) ; 2017: **7** (VIII. Kőzettani és Geokémiai Vándorgyűlés, 12. Téli Ásványtudományi Iskola, MTA Tudományos ülés, ILP MNB ülés); 2018: 1 (MTA Tudományos ülés)

In international conferences or workshops:

2015: **4** (6th workshop on the Neogene of Central and Sout-Eastern European RCMNS Interim Colloquium, Goldschmidt 2015, 26th IUGG General Assembly, 2nd Volcandpark Conference); 2016: **1** (VMSG2016); 2017: **1** (IAVCEI 2017 General Assembly); 2018: **2** (EGU 2018, INQUA–INTAV International Field Conference and Workshop)

Abstracts:

In Hungarian:1 (2015); 1 (2016); 4 (2017)

In English: 4 (2015); 1 (2016); 1 (2017); 2 (2018)

Journal publications:

2015:

Lukács, R., Harangi, S., Bachmann, O., Guillong, M., Danišík, M., Buret, Y., von Quadt, A., Dunkl, I., Fodor, L., Sliwinski, J., Soós, I., Szepesi, J. (2015): Zircon geochronology and geochemistry to constrain the youngest eruption events and magma evolution of the Mid-Miocene ignimbrite flare-up in the Pannonian Basin, eastern central Europe. Contributions to Mineralogy and Petrology, 170(5-6):1-26.

2016:

Petrik, A, Beke, B., Fodor, L., Lukács,R. (2016): Cenozoic structural evolution of the southwestern Bükk Mts. and the southern part of the Darnó Deformation Belt (NE Hungary) Geologica Carpathica 67:(1) pp. 83-104.

2017:

Szepesi, J., Harangi, Sz., Ésik, Zs., Novák, J.T., Lukács, R., Soós, I. (2017): Volcanic Geoheritage and Geotourism Perspectives in Hungary: a Case of an UNESCO World Heritage Site, Tokaj Wine Region Historic Cultural Landscape, Hungary. Geoheritage Volume 9, Issue 3, pp 329–349, doi:10.1007/s12371-016-0205-0

2018:

Lukács R., Guillong M., Sliwinski J., Dunkl I., Bachmann O., Harangi Sz. (2018): LA-ICP-MS U-Pb zircon geochronology data of the Early to Mid-Miocene syn-extensional massive silicic volcanism in the Pannonian Basin (East-Central Europe) Data in Brief Volume 19, August 2018, Pages 506-513; Open Access

Lukács R., Harangi Sz., Guillong M., Bachmann O., Fodor L., Buret Y., Dunkl I., Sliwinski J., von Quadt A., Peytcheva I., Zimmerer M. (2018): Early to Mid-Miocene syn-extensional massive silicic volcanism in the Pannonian Basin (East-Central Europe): Eruption chronology, correlation potential and geodynamic implications Earth-Science Reviews 179: pp. 1-19. Invited Review

Szepesi J., Ésik Zs., Soós I., Novák T. J., Sütő L., Rózsa P., Lukács R., Harangi Sz. (2018): Földtani objektumok értékminősítése: módszertani értékelés a védelem, bemutatás, fenntarthatóság és a geoturisztikai fejlesztések tükrében. Földtani Közlöny, 148/2. (nincs feltüntetve a pályázat)

Manuscript accepted for journal publication:

Petrik, A., Fodor, L., Bereczki, L., Klembala, Zs., Lukács, R., Baranyi, V., Harangi, Sz., Beke, B.: Identification and interpretation of various subvolcanic-volcanic features within a major tectonic zone in the north-eastern part of the Pannonian Basin. Basin Research

Manuscripts under preparation:

Lukács, R., Harangi, Sz., Bachmann, O., Guillong, M., Sliwinski, J., Soós, I., Szepesi, J.: Correlation and magma evolution based on zircon trace element and isotopic compositions: Fingerprints for tracing the distal equivalents of the Miocene silicic ignimbrites of the Northern Pannonian basin.

Lukács, R., Harangi, Sz., Szarvas, I., Pálfy, J, Sztanó, O., Sebe, K., Gál, P., Bachmann, O., Guillong, M., Sliwinski, J.: Distal pyroclastic rock equivalents of the Bükkalja Volcanic Field volcanism in Hungary.

Additional cited reference in this report:

Kelemen, P., Dunkl, I., Csillag, G., Mindszenty, A., von Eynatten, H., Józsa, S. (2017): Tracing multiple resedimentation on an isolated karstified plateau: The bauxite-bearing Miocene red clay of the Southern Bakony Mountains, Hungary. Sedimentary Geology, Volume 358, Pages 84-96.