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$^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of Neogene phreatomagmatic volcanism in the western Pannonian Basin, Hungary

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Abstract

Neogene alkaline basaltic volcanic fields in the western Pannonian Basin, Hungary, including the Bakony–Balaton Highland and the Little Hungarian Plain volcanic fields are the erosional remnants of clusters of small-volume, possibly monogenetic volcanoes. Moderately to strongly eroded maars, tuff rings, scoria cones, and associated lava flows span an age range of ca. 6 Myr as previously determined by the K/Ar method. High resolution $^{40}\text{Ar}/^{39}\text{Ar}$ plateau ages on 18 samples have been obtained to determine the age range for the western Pannonian Basin Neogene intracontinental volcanic province. The new $^{40}\text{Ar}/^{39}\text{Ar}$ age determinations confirm the previously obtained K/Ar ages in the sense that no systematic biases were found between the two data sets. However, our study also serves to illustrate the inherent advantages of the $^{40}\text{Ar}/^{39}\text{Ar}$ technique: greater analytical precision, and internal tests for reliability of the obtained results provide more stringent constraints on reconstructions of the magmatic evolution of the volcanic field. Periods of increased activity with multiple eruptions occurred at ca. 7.95 Ma, 4.10 Ma, 3.80 Ma and 3.00 Ma.

These new results more precisely date remnants of lava lakes or flows that define geomorphological marker horizons, for which the age is significant for interpreting the erosion history of the landscape. The results also demonstrate that during short periods of more intense activity not only were new centers formed but pre-existing centers were rejuvenated.

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1. Introduction

Intracontinental volcanic fields commonly are characterized by low magma supply rates and prolonged activity over periods of millions of years (Walker, 1993; Takada, 1994; Connor et al., 2000). They typically consist of scattered volcanic vents that are often considered to be monogenetic as they apparently never constructed significant composite edifices (Walker, 1993). However, on closer inspection many of the vents do show signs of

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multiple eruption histories (Németh et al., 2003), and their architecture can be complex despite their small size; establishing a time line for individual centers is thus important for understanding their evolution. In addition to the smaller centers, large shield volcanoes and lava flow fields may also occur in these fields (Hasenaka, 1994). Fundamental physical characteristics of volcanic fields include 1) the number, type, eruption styles, sedimentation and erosion history of individual volcanoes (White, 1990; Németh and Martin, 1999a); 2) the timing and frequency of eruptions (Connor et al., 2000); 3) the distribution of volcanoes (Connor et al., 1992); and 4) the relationship of the volcanoes to tectonic features such as basins, faults, and rift zones (Conway et al., 1997). Characterizing such features provides information on magma generation and ascent and will provide a quantitative basis for comparisons among different volcanic fields.

The Neogene western Pannonian volcanic fields were shown during the past decade to have been predominantly phreatomagmatic in eruption style (Németh et al., 2001; Martin and Németh, 2004). Interaction of abundant meteoric water and uprising magma generated explosions that produced the maars and tuff rings. However, there is also evidence for non-explosive, peperite-forming interactions between wet host sediment and intruding, predominantly basanite melt (Martin and Németh, 2007). The resulting craters have been filled by lava in cases where the magma supply was large enough. The timing of the volcanic events in western Hungary has been a concern for a long time (Lóczy, 1913), which has been generally addressed in the last 2 decades by several studies applying the K/Ar technique (Balogh et al., 1982, 1986; Pécskay et al., 1995; Balogh and Pécskay, 2001). This work revealed that the duration of volcanism was ca. 6 Myrs, from about 8 Ma up to 2 Ma. The initiation of volcanism appears to be well constrained at ca 8.0 Ma by several attempts to gain precise K/Ar ages from a maar volcanic complex at Tihany (Balogh and Németh, 2005), but possible episodicity, synchronicity, and the timing of culmination and termination of activity is still under debate. Here, in this paper, we shed new light on these questions by presenting for the first time a set of high precision $^{40}\text{Ar}/^{39}\text{Ar}$ isotope age data from Neogene volcanic rocks of this region.

The primary aim of this study was twofold, 1) to measure the age of samples from selected key locations where the present level of volcanological knowledge is sufficient enough to allow a significant step forward in our understanding of the timing and recurrence rate of the volcanism, and 2) to evaluate the existing K/Ar data set in comparison with the new $^{40}\text{Ar}/^{39}\text{Ar}$ ages.

2. Geological setting

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Neogene intracontinental volcanic fields are present in the Pannonian Basin and consist mostly of alkali basalts and basanites (Downes et al., 1992; Szabó et al., 1992; Embey-Isztin et al., 1993). Volcanism is thought to be related to extensional tectonics, and was shown to have developed along fault lines in the central part of the Pannonian Basin (Jámbor, 1989; Magyar et al., 1999). The volcanic centers in the Pannonian Basin are strongly eroded as the result of basin inversion since the Pliocene, and often only their root zones and feeding channels have been preserved (Conway et al., 1997). By size, inferred eruption mechanisms, distribution pattern, and erosion levels these volcanic fields are considered to be similar to other eroded monogenetic intracontinental volcanic fields such as the Hopi Buttes, Arizona (White, 1991). Volcanic features range from well preserved circular lava capped buttes that mark syn-volcanic paleo-surface levels, to diatremes that indicate locations that are eroded up to hundreds of metres below the syn-volcanic paleosurface (Conway et al., 1997).

In western Hungary, two closely related volcanic fields are the focus of the present study (Fig. 1): the Bakony–Balaton Highland Volcanic Field (BBHVF) and the Little Hungarian Plain Volcanic Field (LHPVF). Though close to one another, the two fields show differences in preserved physical features; phreatomagmatic volcanoes in the northern LHPVF tend to be broader, lensoid landforms and peperites are common in their preserved crater/vent volcanic facies (Martin and Németh, 2005). The depth of magma — water interaction in these volcanoes is inferred to have been less than 300 m below the syn-volcanic paleosurface (Martin and Németh, 2004). The presence of peperites indicates that the host sediment (both siliciclastic and pyroclastic) into which the magma intruded or onto which lava erupted was water-saturated (Martin and Németh, 2005). In contrast, in the BBHVF, especially in the central and eastern part, large numbers of volcanic remnants exhibit features characteristic of magma-water interaction at deeper levels as e.g. in diatremes (Németh et al., 2001). In addition, there are two large shield volcanoes, Kab-hegy and Agát-tető respectively, in the northern part of the BBHVF. The location of volcanic vents is inferred to be related to the distribution of stream filled paleo-valleys as well as to ancient, probably rejuvenated pre-Neogene faults (Németh and Martin, 1999a).

The underlying basement to the volcanic fields in western Hungary largely consists of platform sediments belonging to the Alpine–Carpathian domain, which form a large anticline in the area of the Transdanubian

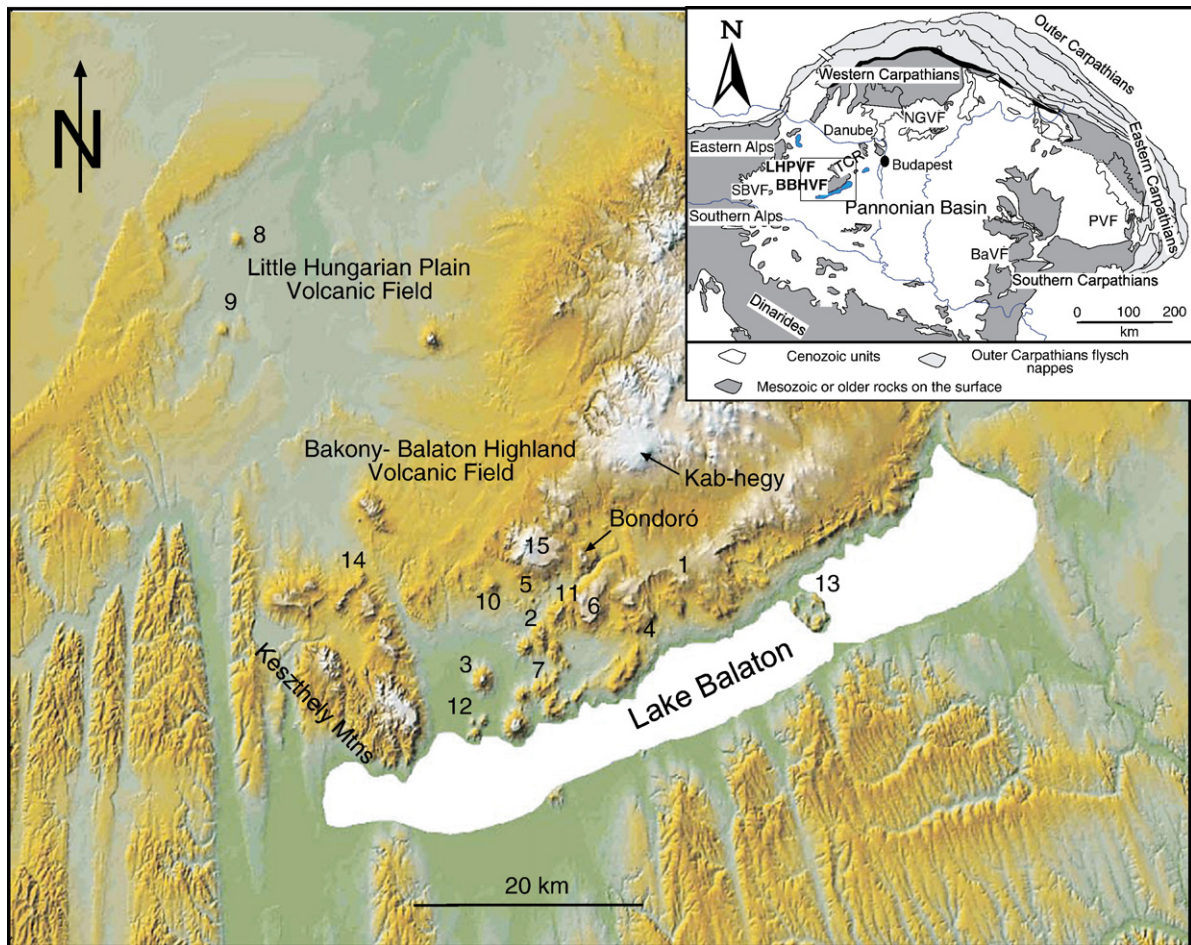


Fig. 1. Simplified geology map of the western Hungarian volcanic fields showing sites from where $^{40}\text{Ar}/^{39}\text{Ar}$ age determinations have been done; (1) Halomhegy scoria cone (HAL1) and lava flow (HAL2), (2) Hajagos maar filling lava lake (HAJ), (3) Szent-György-hegy lava lake (SztGY), (4) Hegyestű plug (HT-6), (5) Hegyesd diatreme (HD10), (6) Fekete-hegy maar crater filling lava lake (FH-4), (7) Tótihegy basanite plug/sill (TW13), (8) Ság-hegy tuff ring (SG2), (9) Kissomlyó tuff ring (KS-1), (10) Haláp maar (HA-1), (11) Füzes-tó scoria cone (FT7), (12) Szigliget diatreme (VAR) and coherent lava flow (SzgD), (13) Tihany Maar Volcanic Complex (TIH), (14) Sümegprága sill and dyke complex (SP), (15) Agár-tető shield volcano (AG1 and AG2).

Central Range (Martin and Németh, 2005). The oldest units consist of a thick package of Silurian schists, Permian terrestrial red sandstones and Alpine-type Mesozoic carbonate platform sediments. During the Neogene, immediately prior to initiation of volcanism, a large lake occupied the Pannonian Basin, the Pannonian Lake (Kázmér, 1990; Magyar et al., 1999) in which a thick sequence of siliciclastic sediments was deposited (Jámbor, 1989; Müller, 1998; Juhász et al., 1999). At the time volcanism began, the area was an alluvial plain (Magyar et al., 1999) on which shallow lakes existed and shallow subaqueous-to-emergent volcanism is inferred on the basis of the textures of pyroclastic rock units as well the common occurrence of peperites (Martin and Németh, 2005, 2007).

On the basis of unconformity-bounded continental sedimentary units in the Neogene stratigraphy of the western Pannonian Basin, three major maximum flooding surfaces have been identified and dated by magnetostratigraphic correlation at 9.0 Ma, 7.3 Ma and around 5.8 Ma (Lantos et al., 1992; Sacchi et al., 1999). The first maximum flooding event correlates with *Congeria czjzeki* fossils in lacustrine beds (Lörenthey, 1900; Müller and Magyar, 1992; Magyar et al., 1999), which mark the Lower Pannonian stage of Lörenthey (1900). After the flooding event, a significant base level drop and subaerial erosion took place around 8.7 Ma (Müller and Magyar, 1992; Sacchi et al., 1999). The second maximum flooding event took place around 7.3 Ma and it is considered to be represented by strata containing *Congeria rhomboidea*

beds (Müller and Magyar, 1992; Sacchi et al., 1997, 1999). A general lowstand and subaerial conditions in the marginal areas is estimated to have occurred around 6 Ma (Sacchi et al., 1999), followed by the last known flooding around 5.3 Ma. On the basis of present knowledge, the ages of volcanic eruptions mostly postdate the latest highstand of the shrinking Pannonian Lake (i.e. younger than 5.3 Ma; Balogh et al., 1982, 1986) with the volcanoes erupted onto an erosion surface (Lóczy, 1913). Precise ages of volcanic rocks, and their correlation with the established eruptive history (subaerial versus shallow subaqueous/emergent) can provide important constraints for reconstruction of the sedimentary and landscape evolution of western Hungary since 9 Ma.

The western Hungarian volcanic fields form the eastern extent of a zone of Neogene intracontinental volcanism in central Europe that formed multiple volcanic fields, including the Massif Central in central France in the west, the Eifel volcanic field in the north and the Slovakian and Hungarian volcanic fields in the east. In western Hungary the formation of the Bakony – Balaton Highland volcanic field and the Little Hungarian Plain volcanic field resulted from 1) deep processes: melt supply from the lithospheric mantle, 2) crustal processes: the Pannonian basin itself was formed by early to mid-Miocene extension, and the volcanic field is situated in the northern block of the Balaton fault zone that is one of the major fault zones controlling the development of the Pannonian basin, and 3) surface processes: the water saturated near surface sediments in the late Miocene and Pliocene were the cause of the explosive character of most of the volcanic events.

3. Analytical techniques

The basalt samples were prepared using standard laboratory techniques (Koppers et al., 2001): following crushing and sieving 250–500 μm fragments were leached in dilute HNO_3 and HF in order to remove alteration phases. Any phenocryst phases present (plagioclase, clinopyroxene and olivine) were routinely removed before packaging ca 250 mg of groundmass in Al-foil packages. Sample packages and ca 5 mg aliquots of laboratory standard sanidine DRA-2 (25.26 Ma, intercalibrated against TCR-1 sanidine at 28.34 Ma; Renne et al., 1998) were sealed in 9 mm diameter quartz glass tubes, with one standard package positioned between every two packages of unknowns.

The irradiation of the tube was carried out for a period of 2 h in a standard 80 mm tall, 25 mm diameter high purity Al sealed tube inserted in a Cd-lined tube in the rotating RODEO poolside facility of the EU-JRC HFR

reactor, Petten, The Netherlands, with the sample capsule positioned in the centre of the neutron field. The neutron flux profile across the reactor is optimized such as to give a negligible flux gradient across the central 12 cm of the Cd-tube. Rotation of the tube during irradiation (60 min^{-1}) helps to minimize the horizontal flux gradient in the tube. The correction factors for the Cd-lined RODEO tube were determined in numerous experiments in our laboratory using high purity Fe doped Ca-silicate and K-silicate glass at $(^{40}\text{Ar}/^{39}\text{Ar})_{\text{K}}$: 0.00183 ± 0.00010 , $(^{39}\text{Ar}/^{37}\text{Ar})_{\text{Ca}}$: 0.000699 ± 0.0000001 , and $(^{36}\text{Ar}/^{37}\text{Ar})_{\text{Ca}}$: 0.000270 ± 0.0000001 .

Upon return to the laboratory, the standard minerals were loaded ca. 4–6 grains per position, (5 replicates for each position) in a Cu sample tray (diameter 66 mm, sample holes 2 mm diameter, 3 mm depth, 185 positions) in a low volume UHV gas sample purification line (Wijbrans et al., 1995) and fused by a laser single fusion technique under full software control. The laser beam, CW argon ion laser with principle lines at 488 nm and 514.5 nm and variable laser power up to 24 W in all lines mode, was focused to a ca. 200 μm spot size, and under software control, the x – y stage is moved in 4 circles increasing in diameter from ca 500 μm to 2000 μm to ensure that all individual crystals are fused using a ca. 15 W laser beam in the experiment. From each sample ca 50 mg was loaded in a Cu sample tray (diameter 66 mm, 22 sample holes of 6 mm diameter, 3 mm deep, and 60° angle to the wall to prevent laser shadows at the bottom of the pan). The rock fragments were spread out evenly in each position in the tray to ensure uniform laser heating. The laser beam was defocused to a ca. 2000 μm spot. The software controlled x – y stage moves the sample holder in a raster pattern (three runs right to left direction followed by three runs perpendicular to the first) under the laser beam to ensure event heating of the whole sample. Laser heating under these parameters lasted for 218 s, followed by 436 s clean time, which was sufficient to admit clean argon gas into the mass spectrometer. The 5 isotopes of argon (m/e : 40–36) and their low mass side baselines (at half mass distance) were measured sequentially by magnet field controlled peak hopping on an MAP 215-50 double focusing noble gas mass spectrometer fitted with a Johnston MM1 SEM detector operated at a relative gain of 500 with respect to the Faraday collector (10^{11} Ohm resistor on the Faraday collector amplifier). The SEM amplifier is fitted with three switchable resistors (10^9 , 10^8 , and 10^7 Ω), that will switch to an appropriate range after the ^{40}Ar beam intensity is measured during the peak centering routine at the beginning of each measurement. The integration time for each beam is variable at 1 s increments. Typical

t1.1 Table 1

t1.2 Summary of $^{40}\text{Ar}/^{39}\text{Ar}$ age data from Neogene western Hungarian volcanic fields (single fusion results of an exploratory series are presented in the left columns; plateau ages and isotope correlation data for the incremental heating experiments in the centre and right columns. Full data tables are available in electronic format

| t1.3 | Single fusion experiments | | | | Incremental heating experiments | | | | | | | |
|-------|---------------------------|----------|--------|---------------|---------------------------------|--|------|--------|------|---------------|--------------------|-------------------|
| t1.4 | | | Age | $\pm 1\sigma$ | Plateau age $\pm 1\sigma$ | MSWD% ^{39}Ar , n steps Inverse | | | | MSWD | K/Ca $\pm 1\sigma$ | |
| | | | | | | Isochron age $\pm 1\sigma$ | | | | | | |
| t1.5 | HAL1 | VU45-A12 | 3297.9 | ± 180.4 | 4.08 | ± 0.05 | 1.78 | 36.19 | 3.87 | ± 0.17 | 1.50 | 0.023 ± 0.000 |
| t1.6 | | | | | | $\pm 1.11\%$ | | 5 | | $\pm 4.47\%$ | | |
| t1.7 | HAL2 | VU45-A14 | 3903.8 | ± 39.7 | 3.82 | ± 0.03 | 1.09 | 86.76 | 3.85 | ± 0.04 | 1.11 | 1.14% ± 0.002 |
| t1.8 | | | | | | $\pm 0.74\%$ | | 9 | | $\pm 0.11\%$ | | |
| t1.9 | HAJ | VU45-A15 | 3803.5 | ± 40.6 | 3.80 | ± 0.02 | 1.91 | 71.71 | 3.74 | ± 0.02 | 0.76 | 0.246 ± 0.004 |
| t1.10 | | | | | | $\pm 0.51\%$ | | 7 | | $\pm 0.65\%$ | | |
| t1.11 | SzgD | VU45-A17 | 3959.8 | ± 47.2 | 4.53 | ± 0.05 | 1.64 | 79.68 | 4.33 | ± 0.09 | 1.01 | 0.120 ± 0.002 |
| t1.12 | | | | | | $\pm 1.01\%$ | | 9 | | $\pm 2.11\%$ | | |
| t1.13 | SztGY | VU45-A18 | 4355.5 | ± 24.6 | 4.22 | ± 0.04 | 1.68 | 86.57 | 4.14 | ± 0.13 | 1.60 | 0.286 ± 0.004 |
| t1.14 | | | | | | $\pm 0.87\%$ | | 9 | | $\pm 3.12\%$ | | |
| t1.15 | HT-6 | VU45-B2 | 7934.2 | ± 47.4 | 7.94 | ± 0.03 | 1.86 | 46.22 | 7.78 | ± 0.07 | 0.37 | 0.075 ± 0.001 |
| t1.16 | | | | | | $\pm 0.40\%$ | | 5 | | $\pm 0.93\%$ | | |
| t1.17 | HD10 | VU45-B3 | 3671.4 | ± 83.2 | 4.12 | ± 0.01 | 2.22 | 95.87 | 3.90 | ± 0.10 | 1.50 | 0.062 ± 0.001 |
| t1.18 | | | | | | $\pm 0.33\%$ | | 10 | | $\pm 2.46\%$ | | |
| t1.19 | FH-4 | VU45-B9 | 3857.9 | ± 21.9 | 3.81 | ± 0.02 | 1.57 | 86.65 | 4.72 | ± 0.03 | 1.39 | 0.279 ± 0.004 |
| t1.20 | | | | | | $\pm 0.49\%$ | | 10 | | $\pm 0.66\%$ | | |
| t1.21 | TW13 | VU45-B11 | 4792.4 | ± 25.3 | 4.74 | ± 0.02 | 1.91 | 90.51 | 4.72 | ± 0.04 | 1.98 | 0.207 ± 0.003 |
| t1.22 | | | | | | $\pm 0.36\%$ | | 9 | | $\pm 0.81\%$ | | |
| t1.23 | SG2 | VU45-B12 | 5543.8 | ± 34.1 | 5.48 | ± 0.01 | 1.49 | 54.04 | 5.32 | ± 0.18 | 0.11 | 0.261 ± 0.004 |
| t1.24 | | | | | | $\pm 0.26\%$ | | 4 | | $\pm 3.42\%$ | | |
| t1.25 | KS-1 | VU45-B14 | 4569.5 | ± 32.0 | 4.63 | ± 0.02 | 2.05 | 71.87 | 4.61 | ± 0.02 | 1.92 | 0.205 ± 0.003 |
| t1.26 | | | | | | $\pm 0.34\%$ | | 8 | | $\pm 0.45\%$ | | |
| t1.27 | HA-1 | VU45-B15 | 3162.2 | ± 23.9 | 3.06 | ± 0.02 | 1.17 | 100.00 | 3.01 | ± 0.03 | 0.60 | 0.276 ± 0.004 |
| t1.28 | | | | | | $\pm 0.51\%$ | | 11 | | $\pm 0.84\%$ | | |
| t1.29 | FT7 | VU45-B17 | 2759.8 | ± 38.7 | 2.61 | ± 0.03 | 1.20 | 91.65 | 2.52 | ± 0.08 | 1.12 | 0.106 ± 0.002 |
| t1.30 | | | | | | $\pm 1.13\%$ | | 10 | | $\pm 3.13\%$ | | |
| t1.31 | VAR | VU45-B18 | 4171.5 | ± 41.7 | 4.08 | ± 0.02 | 0.99 | 81.64 | 3.85 | ± 0.47 | 1.23 | 0.270 ± 0.004 |
| t1.32 | | | | | | $\pm 0.59\%$ | | 5 | | $\pm 12.30\%$ | | |
| t1.33 | AG-1 | VU51-B2 | 2998.1 | ± 27.8 | 3.00 | ± 0.03 | 1.60 | 99.02 | 3.14 | ± 0.06 | 0.98 | 0.100 ± 0.024 |
| t1.34 | | | | $\pm 0.93\%$ | | $\pm 0.93\%$ | | 9 | | $\pm 1.91\%$ | | |
| t1.35 | AG-2 | VU51-B3 | 3692.0 | ± 38.8 | 3.30 | ± 0.03 | 0.51 | 37.59 | 3.30 | ± 0.04 | 0.61 | 0.512 ± 0.046 |
| t1.36 | | | | $\pm 1.05\%$ | | $\pm 0.80\%$ | | 7 | | $\pm 1.11\%$ | | |
| t1.37 | SP1861 | VU51-B4 | 4153.2 | ± 47.8 | 4.15 | ± 0.05 | 0.85 | 98.88 | 3.81 | ± 0.18 | 0.37 | 0.025 ± 0.009 |
| t1.38 | | | | $\pm 1.15\%$ | | $\pm 1.15\%$ | | 9 | | $\pm 4.74\%$ | | |
| t1.39 | TIH | VU51-B6 | 7987.0 | ± 28.1 | 7.96 | ± 0.03 | 0.51 | 75.89 | 8.01 | ± 0.07 | 0.46 | 0.164 ± 0.043 |
| t1.40 | | | | $\pm 0.35\%$ | | $\pm 0.34\%$ | | 5 | | $\pm 0.86\%$ | | |

272 settings are 10 s for ^{40}Ar and ^{39}Ar beams, 6 s for their
 273 baselines, 20 s for ^{36}Ar beams, and 10 for its baselines,
 274 the integration time on the ^{37}Ar beam is kept low (2 s) in
 275 order to avoid excessive increase in radioactive decay
 276 induced noise in the SEM. For data reduction we used the
 277 in-house developed ArArCalc2.2c software package
 278 (Koppers, 2002) (<http://earthref.org/tools/ararcalc/>).
 279 Mass discrimination was measured several times during
 280 the course of this project using our ^{38}Ar -air gas mixture
 281 (full description of our mass discrimination measure-
 282 ment protocol can be found in (Kuiper, 2003). For the
 283 decay constant and the abundance of ^{40}K we used the
 284 values recommended by the IUGS Subcommittee on

Geochronology (Steiger and Jäger, 1977). Using the
 values for flux monitors, decay constant and ^{40}K abun-
 dence discussed in this paragraph in the 2–8 Ma age
 bracket we are aware of a consistent bias of ca 1%
 towards younger ages between our isotopic measure-
 ments and the APTS developed for cyclically bedded
 Neogene sediments (Hilgen et al., 1999; Gradstein et al.,
 2004; Kuiper et al., 2004, 2005).

4. Results

High resolution $^{40}\text{Ar}/^{39}\text{Ar}$ laser incremental heating
 experiments were carried out on 18 samples from 14

296 locations in the western Pannonian volcanic province.
 297 Full data tables, age spectra, K/Ca spectra and isochrons
 298 can be found in a digital background data set (Back-
 299 ground data set: Table 1), descriptions of the sample
 300 sites and dating results can be found in an appendix
 301 (background data set: Appendix). A summary of K/Ar
 302 ages published previously by Balogh and co-workers is
 303 included as Table 2 in the background data supplement.
 304 A summary of $^{40}\text{Ar}/^{39}\text{Ar}$ results is presented in Table 1
 305 and in Fig. 2.

306 All experiments showed good consistent results with,
 307 in most cases, plateaus that meet commonly accepted
 308 reliability criteria. MSWD values were used to define
 309 the plateau segments (Koppers et al., 2001). All ex-
 310 periments yielded plateau segments with MSWDs
 311 indicating that the gas was derived from one isotopically
 312 homogeneous reservoir. For HD10 and KS-1 the
 313 calculated MSWDs were slightly higher than 2.0, as
 314 the result of low individual analytical step uncertainties.
 315 None of the samples showed significant amounts of
 316 excess or inherited ^{40}Ar in the non-radiogenic intercepts
 317 of the normal and inverse isochrons. Nor did any
 318 samples show evidence for profound overprinting
 319 subsequent to deposition, with the exception of sample
 320 VAR (from the Szigliget Vár-hegy pyroclastic succes-
 321 sion) which nevertheless yielded an acceptable plateau
 322 age. Several spectra showed elevated ages in the initial
 323 steps which may either point to loosely bound excess
 324 ^{40}Ar or, alternatively, to recoil loss of ^{39}Ar from fine
 325 grained alteration phases (Koppers, 2002). Several
 326 experiments thus yielded mildly sloping inverse stair-
 327 case spectra, step by step decreasing, still within accept-
 328 able limits forming a plateau, but perhaps indicative of

329 mild alteration and consequent recoil loss over substan-
 330 tial parts of the gas release.

331 From the amounts of ^{39}Ar and ^{37}Ar released during
 332 the experiments some information may be obtained on
 333 the chemical composition of the mineral phases con-
 334 tributing to the spectrum. This effect, as shown in the K/
 335 Ca plots (see supplementary data tables), indicates that in
 336 the groundmass separates used for this study K-rich
 337 mineral phases consistently dominate during the first
 338 half of the experiment whereas towards higher experi-
 339 ment temperatures proportionally more gas is derived
 340 from Ca-rich phases. When the variation in K/Ca is
 341 larger than one order of magnitude, both end member
 342 phases contribute to the plateau age, which suggests that
 343 the K-rich phase observed in the first halves of the
 344 experiments is a primary magmatic phase and not an
 345 alteration product. The exception to this observation is
 346 sample AG2 (from a scoria cone remnant topping the
 347 Agár-tető shield volcano) where the phase enriched in Ca
 348 actually has a slightly, but in terms of finding a plateau,
 349 significantly increased age with respect to the plateau
 350 segment. The radiogenic component of the argon ranges
 351 from less than 10% to ca 80%. The low amounts of
 352 radiogenic argon typically found in the samples with low
 353 K/Ca is reflected in their proportionally larger analytical
 354 uncertainties (e.g. sample VAR).

5. Discussion

355 The new $^{40}\text{Ar}/^{39}\text{Ar}$ ages show that volcanism oc-
 356 curred in two broad periods: the first period is confined to
 357 two eruption centres formed along the north shore of
 358 Lake Balaton, Tihany and Hegyes-tű (Fig. 2, Episode I).
 359

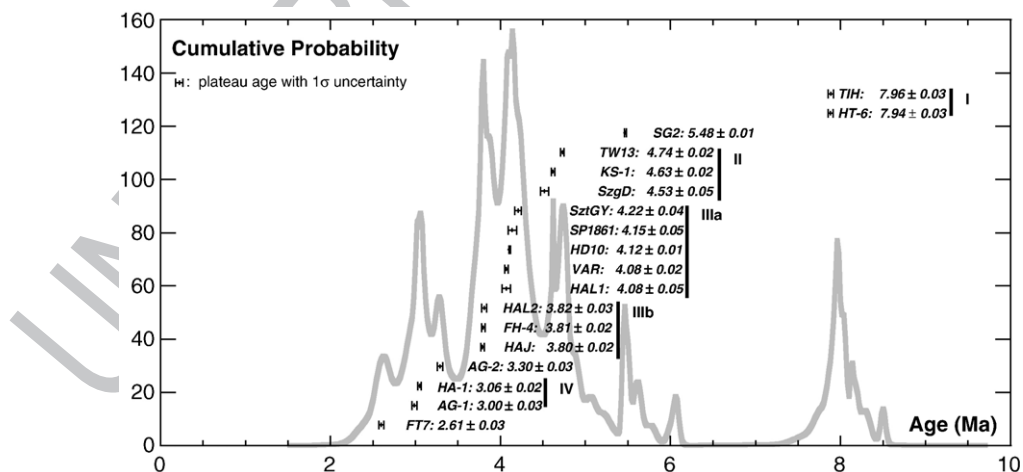


Fig. 2. Cumulative probability diagram showing all $^{40}\text{Ar}/^{39}\text{Ar}$ age information obtained for this study. All individual step ages and their 1σ uncertainties have been used to construct the cumulative probability curve in the diagram. Plateau ages and their 1σ uncertainty intervals are indicated as bars to the left of the individual ages. Age groups (Episodes I, II, III and IV) are identified with Roman numerals.

360 The age results for these two centres, 7.94 ± 0.03 Ma
 361 and 7.96 ± 0.03 Ma are identical suggesting that we
 362 are dealing with two surface exposures of rocks from the
 363 same eruption. The other 16 samples (Fig. 3) define the
 364 second broad period of activity that formed of the
 365 volcanic field with eruptions starting ca. 5.5 Myr ago and
 366 reaching a culmination around 4.0 Ma (Fig. 2) with
 367 activity recorded at Halom-hegy: 4.08, 3.82 Ma, Haja-

gos: 3.80 Ma (Fig. 3a), Hegyesd: 4.12 Ma, Fekete-hegy 368
 lava field: 3.81 Ma, the Szigliget diatreme Vár-hegy 369
 pyroclastic sequence: 4.08 Ma, and the Sümegprága sill: 370
 4.15 Ma). This second broad period ended ca 2.6 Myr 371
 ago. 372

In addition to the broad division into two periods, the 373
 first centred around 8.0 Ma and the second centred 374
 around 4.0 Ma, it was noted that eruptions in different 375

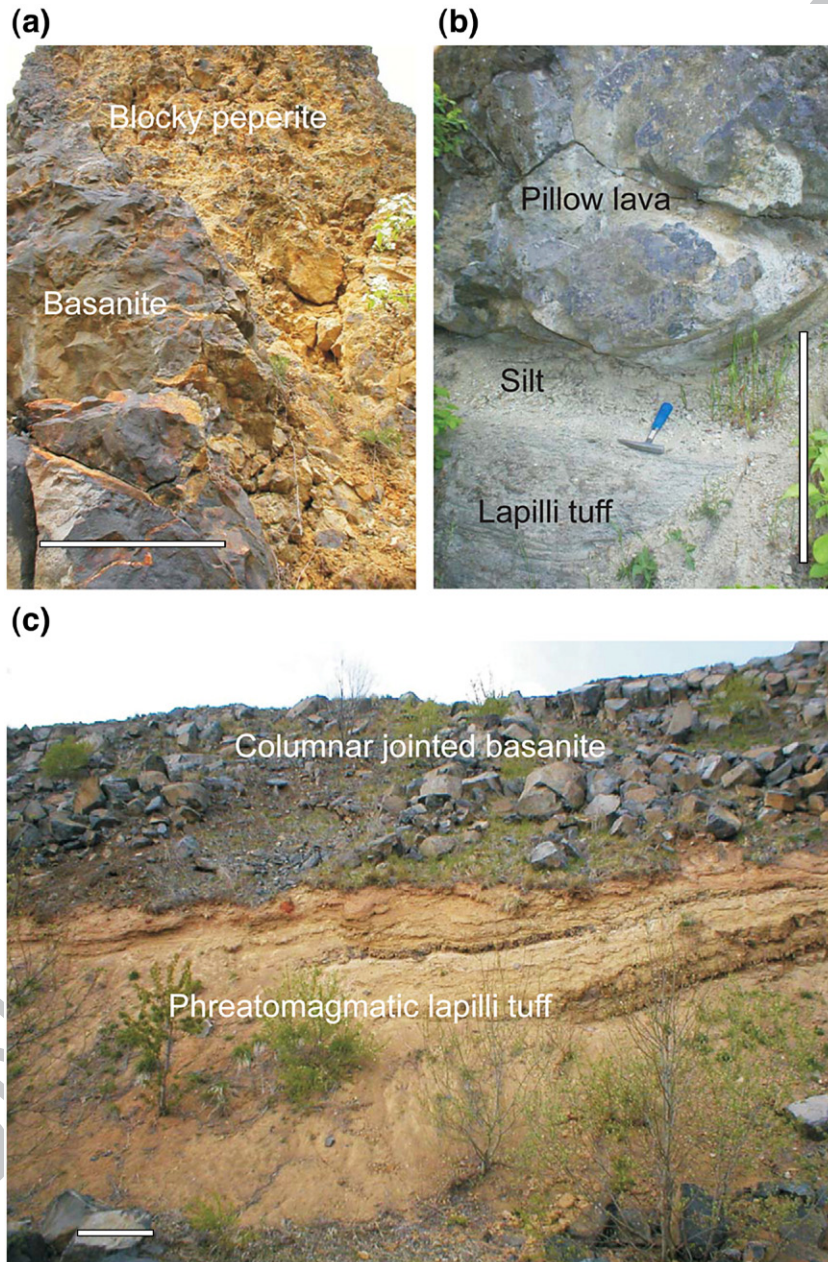


Fig. 3. Measured samples from a) dated blocky peperite from Hajagos (Location 2). Dark angular clasts are the basanite hosted in fine sediment; b) Kissomlyó (Location 9) pyroclastic unit overlain by siliciclastic beds invaded by the dated lava, c) columnar jointed basanite overlain the tuff ring units at Haláp maar (Location 10). White bars represent 1 m on each figure.

376 volcanic centres often yielded age results that were
377 indistinguishable. This observation forms the basis
378 for dividing volcanic activity of the field into 5 distinct
379 episodes (I, II, IIIa, IIIb and IV). These episodes are
380 defined as periods of activity yielding tightly clustering
381 ages, often within the individual 2 sigma uncertainties of
382 the plateau age results. Three ages, that of SG2 ($5.48 \pm$
383 0.01 Ma), AG-2 (3.30 ± 0.03 Ma, and FT7 ($2.61 \pm$
384 0.03 Ma) have not been shown as occurring in different
385 centres.

386 The oldest ages from our $^{40}\text{Ar}/^{39}\text{Ar}$ geochronol-
387 ogy were derived from a basanite plug of Hegyes-tű
388 (7.94 Ma) and the Tihany volcano (7.96 Ma), together
389 defining Episode I. These ages are in excellent
390 agreement with the 7.92 Ma K/Ar age of the Tihany
391 maar volcanic complex (Balogh and Németh, 2006),
392 and represent the oldest ages from the western
393 Hungarian alkaline basaltic volcanic fields. These ages
394 fall in the time between the maximum highstands of the
395 Pannonian Lake at 9.0 Ma (msf-2) and 7.3 Ma (msf-3)
396 (Sacchi et al., 1999). A lowstand characterized by ero-
397 sion and widely exposed marginal lake banks is inferred
398 to have developed around 8.7 Ma ago (Sacchi et al.,
399 1999). The ages of Hegyes-tű and Tihany (this work,
400 Balogh and Németh, 2005) suggest, that these volcanoes
401 erupted in the phreatic zone of the Pannonian Lake, near
402 to its shoreline, where water to sustain phreatomagmat-
403 ism was likely available from the large water mass of
404 the nearby lake (Németh et al., 2001). The likely paleo-
405 geomorphological scenario would be similar to that of
406 the Newer Volcanics in Victoria, Australia, or the Recent
407 Ukinrek Maars formed in the 1977's in Alaskan Pen-
408 insula, Alaska where volcanic fields have developed in a
409 near shore environment (Self et al., 1980; Johnson,
410 1989; Jones et al., 2001).

411 The main activity during the younger period occurred
412 around 4.0 Ma (Episode III). On the basis of the plateau
413 results this group might be subdivided into an older
414 sub group (episode IIIa) and a younger subgroup (epi-
415 sode IIIb). The isochron results would suggest that all
416 these eruptive products belong to one single group. The
417 Szigliget diatreme age is relatively poorly determined,
418 due to a low level of radiogenic $^{40}\text{Ar}^*$ and consequent
419 larger error in the dating results. The significance of the
420 similarity of these ages is, that the large lava field of the
421 Fekete-hegy can be viewed as a marker horizon, a ca.
422 3.81 Myr old paleosurface preserved by the lava. The
423 Fekete-hegy lava flow has a contact with pyroclastic
424 rock units at an altitude of ~ 340 m a.s.l., similar to that at
425 Hajagos (~ 320 m level contact with pyroclastic rocks),
426 and to the altitude of the uppermost deposits of the pre-
427 volcanic siliciclastic succession. Taking these values into

account, and inferring a fairly uniform paleosurface over 428
the area of the field would imply that the topmost ex- 429
posures of the Hegyesd and Szigliget diatremes (~ 260 m 430
and ~ 220 m a.s.l., respectively) still would be around 431
80–100 m below the syn-volcanic paleosurface. This 432
estimate is in good agreement with volcanological 433
observations and the interpretation that these two sites 434
represent exposed diatremes (conduits of former phrea- 435
tomagmatic volcanoes). The total thickness of pre- 436
volcanic, mostly Pannonian (Upper Miocene) sand and 437
silt eroded since these volcanoes erupted 3.8–4.2 Myr 438
ago would be around 200–250 m, implying a 50–65 m/ 439
Myr long term averaged erosion rate for these sites. Szent 440
György-hegy with an age of 4.22 ± 0.4 Ma is the oldest 441
centre with activity during this period. These estimates 442
are in the same range as those inferred previously on the 443
basis of volcanic facies analyses and published K/Ar 444
ages (Németh and Martin, 1999a). 445

446 The volcanic vents belonging to Episode III are as- 446
sociated with phreatomagmatic pyroclastic units in- 447
terpreted as evidence that the magma interacted with 448
abundant water (Németh and Martin, 1999b). The tex- 449
tural characteristics of the pyroclastic sequences indicate 450
that phreatomagmatic explosions took place below a 451
subareal paleosurface, i.e. not under lacustrine condi- 452
tions (Németh and Martin, 1999b). The great variety of 453
peperite at Hajagos (Fig. 3A) (Martin and Németh, 2007) 454
suggests, however, that sufficient amounts of water were 455
present in a near-surface aquifer to fill the maar basins 456
created by the explosive eruptions. In these water-filled 457
basins, newly erupted basanite melt interacted with the 458
water saturated wall-rock, crater wall, and pre-volcanic 459
mud and silt to form various peperites (Fig. 3a) (Martin 460
and Németh, 2007). The age of Fekete-hegy and as- 461
sociated sites corresponds well with the proposed time at 462
which the Pannonian Lake dried up (Sacchi et al., 1997, 463
1999; Magyar et al., 1999; Sacchi and Horváth, 2002), 464
and thus is consistent with the observation of subareal 465
magmatism in combination with a water-saturated, near 466
surface, aquifer. Conditions at this time were still sub- 467
stantially wetter than present day conditions in the area. 468

469 A group of volcanoes, designated as belonging to 469
Episode II is slightly older than the Fekete-hegy and 470
associated sites on the basis of the $^{40}\text{Ar}/^{39}\text{Ar}$ ages 471
grouping around 4.5 to 4.8 Ma (Szigliget lava: 4.53 Ma, 472
Kissomlyó: 4.63 Ma and Tóti-hegy: 4.74 Ma: Fig. 2, 473
Episode II). Of this group the Szigliget lava sample 474
should be viewed with some caution. The field 475
relationships between the Szigliget pyroclastic sequence 476
(4.08 Ma) and the coherent lava body (4.53 Ma) are 477
unclear. An intrusive contact of the lava was proposed 478
(Borsy et al., 1986) because of its oblique, non-uniform 479

480 thickness and because both the underlying and overlying
481 rock is pyroclastic rock with very similar textural
482 features. However, the new age data make this interpretation
483 problematic, and instead indicate a ‘normal’
484 layer cake stratigraphy, with an older lava flow overlain
485 by a younger phreatomagmatic pyroclastic succession.
486 Alternatively, Szigliget may represent an erosional
487 remnant of a nested diatreme. In this reconstruction,
488 the age data derived from the pyroclastic rocks and the
489 coherent lava body document two different phreatomagmatic
490 events which occurred about 0.5 Myr apart. The
491 coherent lava body and its host pyroclastic unit in this
492 interpretation should belong to an older diatreme, within
493 which a new diatreme developed. Similar nested
494 diatremes are not unknown, especially from kimberlite
495 fields (Skinner and Marsh, 2004), and, therefore, the new
496 age dating suggests that further research on Szigliget
497 with aimed at understanding its volcanic evolution, is
498 required. One should be cautioned however that the
499 Szigliget samples, especially the cauliflower bomb sample
500 from the capping pyroclastic unit, are from basalt that
501 is particularly low in potassium and hence had a very low
502 enrichment in radiogenic ^{40}Ar . Therefore the analytical
503 uncertainty of its ages is large and, thus, the two ages
504 might still belong to the same event.

505 The $^{40}\text{Ar}/^{39}\text{Ar}$ ages of Szent György-hegy: Kissomlyó
506 and Tóti-hegy seem to indicate an eruptive period from
507 4.2 to 4.8 Ma, which overlaps in time with the period
508 when the Pannonian Lake progressively decreased in size
509 (Sacchi et al., 1999). The age of the lava lake infilling the
510 Kissomlyó tuff ring (Fig. 3b) is 4.63 Ma by the $^{40}\text{Ar}/^{39}\text{Ar}$
511 method. This age is significantly younger than that of the
512 nearby (5.48 Ma) Ság-hegy lava, and, therefore, assuming
513 coeval initiation of volcanism in the Kissomlyó–Ság-
514 hegy area, it can be interpreted as the age of a lava which
515 erupted from the same volcano that produced the
516 Kissomlyó tuff ring. Although all these volcanoes belonging
517 to the 4.2–4.8 Ma period erupted in subaerial conditions,
518 the widespread evidence of phreatomagmatism is considered
519 strong evidence for the abundance of water in the rocks near
520 the Earth’s surface at this time. The presence of peperite,
521 intra-crater lacustrine sediments, and glassy volcanic
522 textures may reflect surface water involvement in the
523 development of the Kissomlyó volcano and suggest that
524 shallow (few metres) standing water bodies may have
525 developed from time to time on the large flat plain of
526 western Hungary (Martin and Németh, 2005).

527 The oldest age (5.48 Ma) for the volcanic field in the
528 younger age group was derived from a peperitic sill from
529 Ság-hegy. This age is correlated with the last lowstand of
530 the Pannonian Lake, however, the pyroclastic succession
531 and intrusive bodies of Ság-hegy clearly demonstrate

532 that they developed in a wet environment. From this
533 observation we argue that after the Pannonian Lake
534 ceased to exist, the first few 100s of metres of the
535 stratigraphy remained water saturated for several millions of
536 years. Thus, after the retreat of the Pannonian Lake, the
537 resultant alluvial plain most likely was littered with small
538 alluvial lakes reflecting a generally high water table
539 and fluctuating in extent with seasonal and climatic
540 variations.

541 The youngest $^{40}\text{Ar}/^{39}\text{Ar}$ ages were found for the
542 Agár-tető shield volcano (AG1: 3.00 Ma), the Haláp tuff
543 ring (3.06 Ma) (Fig. 2, Episode IV) and the Füzés-tó
544 scoria cone (2.61 Ma). The relatively young ages of these
545 localities indicate that their morphology may partly
546 preserve their original volcanic structure. At Haláp, the
547 dated lava flow caps the phreatomagmatic pyroclastic
548 sequence of a tuff ring (Fig. 3c). The lava flow and the
549 pyroclastic sequences have a peperitic contact suggesting
550 that the tephra ring must have been water saturated,
551 therefore, a water-filled crater is inferred. At Haláp no
552 original volcanic landform can be recognized. At Füzés-
553 tó the young age is supported by its well-preserved
554 central depression filled with ballistic bombs and lava
555 spatter indicating that its crater is still intact and un-
556 breached. It is notable that after 2.61 Myr of erosion
557 Füzés-tó still has kept its form, which suggests slow
558 erosion rates and/or that local factors prevented ex-
559 cessive erosion. A young K/Ar age of 2.3 Ma has been
560 measured from Bondoró (Fig. 1), a volcano that is similar
561 to Füzés-tó; however, its crater has been breached
562 (Embey-Isztin, 1993). A similar young age has also been
563 derived from Agár-tető, a capping scoria cone remnant,
564 giving an age of 2.98 Ma by the K/Ar method (Balogh
565 et al., 1982). It seems that the closing stage of the
566 volcanism in western Hungary was around 2.5–2 Ma.

567 In terms of magmatic processes the Western Hungarian
568 volcanic fields are characterized by several episodes
569 during which (near-) synchronous eruptions occurred
570 at multiple centres. This observation is interpreted as
571 evidence for a discrete number of melt emplacement
572 events during which melt generated in the sublitho-
573 spheric mantle was emplaced into the crust. The amounts
574 of magma were sufficient to feed several edifices, but not
575 enough to sustain prolonged magmatism at individual
576 edifices. Although we have identified discrete episodes
577 of magmatism, there is no evidence for periodicity in the
578 data. i.e. from our data we cannot deduce that magmatic
579 events occurred with a predictable frequency: the time
580 span between the onset of magmatism at 7.97 Ma and the
581 second event is 2.5 Myr, the period between the second
582 and third episode is ca 700 000 yr, and between the
583 second and third episodes between 4.65 Ma and 4.10 Ma

584 was 550 000 yr, and between the final episodes between
585 3.80 and 3.00 Ma was ca. 800 000 yr.

586 Several authors have suggested that there is a relation
587 between magmatism and basin extension in the Pannoni-
588 an Basin (Horváth, 1993). The main phase of basin
589 extension in the Pannonian Basin, however, predates the
590 development of the West Hungarian volcanic fields. The
591 onset of magmatism at ca 7.95 Ma in fact occurred
592 during a period of relative quiescence in the basin evolu-
593 tion, and the main phase of magmatism around 4.0 Ma
594 coincides with the onset of basin inversion (Cloetingh
595 et al., 2005; Fodor et al., 2005). While basin inversion
596 was probably responsible for the disappearance of the
597 Pannonian lake in the early Pliocene, there is no clear
598 evidence that it caused the episodes of mantle melting
599 recorded in the volcanic fields of western Hungary.
600 During the early stages of inversion the environment was
601 still wet enough to cause the phreatomagmatic features in
602 the volcanic field, but it is probably significant that one
603 of the shield volcanoes in the area, Agát-tető, is in fact
604 one of the youngest features in the field, and formed after
605 basin inversion had largely dried out the area.

606 6. Conclusion

607 When comparing the existing data set of conventional
608 K/Ar ages with new high resolution $^{40}\text{Ar}/^{39}\text{Ar}$ ages for
609 the volcanism in the western Hungarian alkaline basaltic,
610 intracontinental volcanic fields, we may conclude that
611 the two methods yielded consistent results, provided that
612 the samples are simple groundmass samples with limited
613 alteration and limited excess ^{40}Ar or extraneous ^{40}Ar
614 contained in phenocrysts. The similarity has confirmed
615 that in an absolute sense the timing of the Neogene
616 volcanic events inferred for the Bakony–Balaton and
617 Little Hungarian Plain volcanic fields is correct.
618 However, in addition, we demonstrate the potential of
619 $^{40}\text{Ar}/^{39}\text{Ar}$ dating for establishing volcanic stratigraphies
620 for individual centres. The significant difference be-
621 tween the two methods is the analytical uncertainty,
622 which is an order of magnitude less for $^{40}\text{Ar}/^{39}\text{Ar}$ dating,
623 and the more consistent check for sample homogeneity.
624 However, as we are dealing here with the products
625 of explosive volcanism, some of the material used for
626 dating was highly fragmented during formation: some of
627 the fragments can easily be recognized as bombs formed
628 upon eruption, but other fragments particularly in
629 diatremes and scoria cones cannot easily be character-
630 ized by morphology. In such cases it may not be possible
631 to distinguish syn-extrusive bomb fragments from
632 shattered intrusions from deeper down in the plumbing
633 system. Thus, the real geological problems may cause a

larger range in expected ages and thus the increased
precision of the $^{40}\text{Ar}/^{39}\text{Ar}$ method also should be com-
plemented with more and more focused field research in
order to interpret the isotopic results.

The $^{40}\text{Ar}/^{39}\text{Ar}$ ages confirm that:

- (1) Volcanic activity peaked around 4 million years
ago during perhaps 2 periods of intensified
activity that affected several centers. The older
Tihany–Hegyes-tű period at 7.95 Ma was of more
limited importance, both in areal extent and in
volume of magmatism.
- (2) Volcanism occurred near-synchronously at multi-
ple locations at four times during the history of the
volcanic field: first at ~ 7.95 Ma ($n=2$, at Tihany
and Hegyes-tű), at ~ 4.1 Ma ($n=5$, at Halom-
hegy, Szent György-hegy, Hegyesd, Vár-hegy,
and Sümegprága), at ~ 3.8 Ma ($n=3$, at Halom-
hegy, Hajagos, and Fekete-hegy), and at 3.0 Ma
($n=2$, at Agár-tető, and Haláp).
- (3) There are no clear spatial patterns in the
distribution and timing of volcanism in western
Hungary, There may have been though a slight
east to west shift in the location of vents with
time.
- (4) The very low analytical uncertainties of the
 $^{40}\text{Ar}/^{39}\text{Ar}$ dates allow us to distinguish volcanic
events at closely spaced centres to provide better
understanding of rejuvenation of volcanic eruption
centres at the same place (Kissomlyó vs. Ság-
hegy), and may also be used with success to
confirm more prolonged activity at individual
sites, and thus may cast doubt on whether this type
of volcanism is truly ‘monogenetic’.
- (5) The dated volcanoes erupted at a time of lowstand
in the nearby Pannonian Lake, and despite the
abundant evidence to support a water-rich erup-
tive environment (Ság-hegy, Kissomlyó, Tihany)
these volcanoes are inferred to have erupted in a
subaerial phreatic zone adjacent to the lake itself.

7. Uncited references

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Webb et al., 2004

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695 Appendix A. Supplementary data

696 Supplementary data associated with this article
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$^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of Neogene phreatomagmatic volcanism 3 in the western Pannonian Basin, Hungary

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