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Journal of volcanology and geothermal research

Journal of Volcanology and Geothermal Research xx (2007) xxx-xxx

www.elsevier.com/locate/jvolgeores

⁴⁰Ar/³⁹Ar geochronology of Neogene phreatomagmatic volcanism in the western Pannonian Basin, Hungary

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Received 25 June 2006; received in revised form 8 March 2007; accepted 10 May 2007

11 Abstract

Neogene alkaline basaltic volcanic fields in the western Pannonian Basin, Hungary, including the Bakony-Balaton Highland and 12the Little Hungarian Plain volcanic fields are the erosional remnants of clusters of small-volume, possibly monogenetic volcanoes. 13 Moderately to strongly eroded maars, tuff rings, scoria cones, and associated lava flows span an age range of ca. 6 Myr as previously 14 determined by the K/Ar method. High resolution ⁴⁰Ar/³⁹Ar plateau ages on 18 samples have been obtained to determine the age range 15for the western Pannonian Basin Neogene intracontinental volcanic province. The new ⁴⁰Ar/³⁹Ar age determinations confirm the 16 previously obtained K/Ar ages in the sense that no systematic biases were found between the two data sets. However, our study also 17 serves to illustrate the inherent advantages of the ⁴⁰Ar/³⁹Ar technique: greater analytical precision, and internal tests for reliability of 18 the obtained results provide more stringent constraints on reconstructions of the magmatic evolution of the volcanic field. Periods of 1920increased 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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26 Keywords: phreatomagmatic; pyroclastic; basanite; monogenetic; scoria; ⁴⁰Ar/³⁹Ar geochronology; peperite; erosion

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0377-0273/\$ - see front matter @ 2007 Published by Elsevier B.V. doi:10.1016/j.jvolgeores.2007.05.009

1. Introduction

Intracontinental volcanic fields commonly are charac- 29 terized by low magma supply rates and prolonged activity 30 over periods of millions of years (Walker, 1993; Takada, 31 1994; Connor et al., 2000). They typically consist of 32 scattered volcanic vents that are often considered to be 33 monogenetic as they apparently never constructed sig- 34 nificant composite edifices (Walker, 1993). However, 35 on closer inspection many of the vents do show signs of 36

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multiple eruption histories (Németh et al., 2003), and their 37 architecture can be complex despite their small size; es-38 tablishing a time line for individual centers is thus im-39 portant for understanding their evolution. In addition to 40 the smaller centers, large shield volcanoes and lava flow 41 fields may also occur in these fields (Hasenaka, 1994). 42Fundamental physical characteristics of volcanic fields 43 include 1) the number, type, eruption styles, sedimenta-44 tion and erosion history of individual volcanoes (White, 451990; Németh and Martin, 1999a); 2) the timing and 46 frequency of eruptions (Connor et al., 2000); 3) the dis-47 tribution of volcanoes (Connor et al., 1992); and 4) the 48 49 relationship of the volcanoes to tectonic features such as basins, faults, and rift zones (Conway et al., 1997). Char-50acterizing such features provides information on magma 51generation and ascent and will provide a quantitative basis 52for comparisons among different volcanic fields. 53

54The Neogene western Pannonian volcanic fields were 55shown during the past decade to have been predominantly phreatomagmatic in eruption style (Németh et al., 2001; 56Martin and Németh, 2004). Interaction of abundant me-57teoric water and uprising magma generated explosions 58 that produced the maars and tuff rings. However, there is 59 60 also evidence for non-explosive, peperite-forming interactions between wet host sediment and intruding, pre-61 dominantly basanite melt (Martin and Németh, 2007). 62 The resulting craters have been filled by lava in cases 63 where the magma supply was large enough. The timing 64 65 of the volcanic events in western Hungary has been a concern for a long time (Lóczy, 1913), which has been 66 generally addressed in the last 2 decades by several 67 studies applying the K/Ar technique (Balogh et al., 1982, 68 1986; Pécskay et al., 1995; Balogh and Pécskay, 2001). 69 This work revealed that the duration of volcanism was ca. $\overline{70}$ 6 Myrs, from about 8 Ma up to 2 Ma. The initiation of 71 volcanism appears to be well constrained at ca 8.0 Ma by 72several attempts to gain precise K/Ar ages from a maar 73 volcanic complex at Tihany (Balogh and Németh, 2005), 74 but possible episodicity, synchroneiety, and the timing of 75culmination and termination of activity is still under de-76 bate. Here, in this paper, we shed new light on these 77 questions by presenting for the first time a set of high 78 precision ⁴⁰Ar/³⁹Ar isotope age data from Neogene vol-79canic rocks of this region. 80

The primary aim of this study was twofold, 1) to 81 measure the age of samples from selected key 82 locations where the present level of volcanological 83 knowledge is sufficient enough to allow a significant 84 step forward in our understanding of the timing and 85 recurrence rate of the volcanism, and 2) to evaluate the 86 existing K/Ar data set in comparison with the new 87 40 Ar/ 39 Ar ages. 88

2. Geological setting

Neogene intracontinental volcanic fields are present 90 in the Pannonian Basin and consist mostly of alkali 91 basalts and basanites Downes et al., 1992; Szabó et al., 92 1992; Embey-Isztin et al., 1993). Volcanism is thought 93 to be related to extensional tectonics, and was shown to 94 have developed along fault lines in the central part of the 95 Pannonian Basin (Jámbor, 1989; Magyar et al., 1999). 96 The volcanic centers in the Pannonian Basin are strongly 97 eroded as the result of basin inversion since the Pliocene, 98 and often only their root zones and feeding channels 99 have been preserved (Conway et al., 1997). By size, 100 inferred eruption mechanisms, distribution pattern, and 101 erosion levels these volcanic fields are considered to be 102 similar to other eroded monogenetic intracontinental 103 volcanic fields such as the Hopi Buttes, Arizona (White, 104 1991). Volcanic features range from well preserved cir- 105 cular lava capped buttes that mark syn-volcanic paleo- 106 surface levels, to diatremes that indicate locations that 107 are eroded up to hundreds of metres below the syn- 108 volcanic paleosurface (Conway et al., 1997). 109

In western Hungary, two closely related volcanic 110 fields are the focus of the present study (Fig. 1): the 111 Bakony-Balaton Highland Volcanic Field (BBHVF) 112 and the Little Hungarian Plain Volcanic Field (LHPVF). 113 Though close to one another, the two fields show dif- 114 ferences in preserved physical features; phreatomag- 115 matic volcanoes in the northern LHPVF tend to be 116 broader, lensoid landforms and peperites are common in 117 their preserved crater/vent volcanic facies (Martin and 118 Németh, 2005). The depth of magma — water interac- 119 tion in these volcanoes is inferred to have been less than 120 300 m below the syn-volcanic paleosurface (Martin and 121 Németh, 2004). The presence of peperites indicates that 122 the host sediment (both siliciclastic and pyroclastic) into 123 which the magma intruded or onto which lava erupted 124 was water-saturated (Martin and Németh, 2005). In con- 125 trast, in the BBHVF, especially in the central and eastern 126 part, large numbers of volcanic remnants exhibit features 127 characteristic of magma-water interaction at deeper 128 levels as e.g. in diatremes (Németh et al., 2001). In 129 addition, there are two large shield volcanoes, Kab-hegy 130 and Agát-tető respectively, in the northern part of the 131 BBHVF. The location of volcanic vents is inferred to be 132 related to the distribution of stream filled paleo-valleys 133 as well as to ancient, probably rejuvenated pre-Neogene 134 faults (Németh and Martin, 1999a). 135

The underlying basement to the volcanic fields in 136 western Hungary largely consists of platform sediments 137 belonging to the Alpine–Carpathian domain, which 138 form a large anticline in the area of the Transdanubian 139

Please cite this article as: Wijbrans, J. et al. ⁴⁰Ar/³⁹Ar geochronology of Neogene phreatomagmatic volcanism in the western Pannonian Basin, Hungary. Journal of Volcanology and Geothermal Research (2007), doi:10.1016/j.jvolgeores.2007.05.009

89

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J. Wijbrans et al. / Journal of Volcanology and Geothermal Research xx (2007) xxx-xxx



Fig. 1. Simplified geology map of the western Hungarian volcanic fields showing sites from where ⁴⁰Ar/³⁹Ar 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) Hegyestû 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 140 oldest units consist of a thick package of Silurian 141 schists, Permian terrestrial red sandstones and Alpine-142 type Mesozoic carbonate platform sediments. During 143 the Neogene, immediately prior to initiation of vol-144 canism, a large lake occupied the Pannonian Basin, the 145Pannonian Lake (Kázmér, 1990; Magyar et al., 1999) in 146which a thick sequence of siliciclastic sediments was 147 deposited (Jámbor, 1989; Müller, 1998; Juhász et al., 148 1999). At the time volcanism began, the area was an 149alluvial plain (Magyar et al., 1999) on which shallow 150lakes existed and shallow subaqueous-to-emergent 151volcanism is inferred on the basis of the textures of 152pyroclastic rock units as well the common occurrence 153of peperites (Martin and Németh, 2005, 2007). 154

On the basis of unconformity-bounded continental 155 sedimentary units in the Neogene stratigraphy of the 156 western Pannonian Basin, three major maximum flooding 157 surfaces have been identified and dated by magnetostrati- 158 graphic correlation at 9.0 Ma, 7.3 Ma and around 5.8 Ma 159 (Lantos et al., 1992; Sacchi et al., 1999). The first max- 160 imum flooding event correlates with *Congeria czjzeki* 161 fossils in lacustrine beds (Lörenthey, 1900; Müller and 162 Magyar, 1992; Magyar et al., 1999), which mark the 163 Lower Pannonian stage of Lörenthey (1900). After the 164 flooding event, a significant base level drop and subaerial 165 erosion took place around 8.7 Ma (Müller and Magyar, 166 1992; Sacchi et al., 1999). The second maximum flooding 167 event took place around 7.3 Ma and it is considered to be 168 represented by strata containing *Congeria rhomboidea* 169

beds (Müller and Magyar, 1992; Sacchi et al., 1997, 1701999). A general lowstand and subaerial conditions in the 171 marginal areas is estimated to have occurred around 6 Ma 172(Sacchi et al., 1999), followed by the last known flooding 173around 5.3 Ma. On the basis of present knowledge, the 174 ages of volcanic eruptions mostly postdate the latest 175highstand of the shrinking Pannonian Lake (i.e. younger 176 than 5.3 Ma; Balogh et al., 1982, 1986) with the volcanoes 177erupted onto an erosion surface (Lóczy, 1913). Precise 178 ages of volcanic rocks, and their correlation with the 179 established eruptive history (subaerial versus shallow 180 subaqueous/emergent) can provide important constraints 181 182 for reconstruction of the sedimentary and landscape evolution of western Hungary since 9 Ma. 183

The western Hungarian volcanic fields form the 184 eastern extent of a zone of Neogene intracontinental 185 volcanism in central Europe that formed multiple 186 volcanic fields, including the Massif Central in central 187 France in the west, the Eifel volcanic field in the north 188 and the Slovakian and Hungarian volcanic fields in the 189 east. In western Hungary the formation of the Bakony -190Balaton Highland volcanic field and the Little Hungarian 191 Plain volcanic field resulted from 1) deep processes: melt 192 193 supply from the lithospheric mantle, 2) crustal processes: the Pannonian basin itself was formed by early to mid-194 Miocene extension, and the volcanic field is situated in 195the northern block of the Balaton fault zone that is one of 196 the major fault zones controlling the development of the 197 198 Pannonian basin, and 3) surface processes: the water saturated near surface sediments in the late Miocene and 199 Pliocene were the cause of the explosive character of 200most of the volcanic events. 201

202 3. Analytical techniques

The basalt samples were prepared using standard 203 laboratory techniques (Koppers et al., 2001): following 204 crushing and sieving 250-500 µm fragments were 205leached in dilute HNO₃ and HF in order to remove 206 alteration phases. Any phenocryst phases present 207(plagioclase, clinopyroxene and olivine) were routinely 208 removed before packaging ca 250 mg of groundmass in 209 Al-foil packages. Sample packages and ca 5 mg aliquots 210 of laboratory standard sanidine DRA-2 (25.26 Ma, in-211 tercalibrated against TCR-1 sanidine at 28.34 Ma; Renne 212 et al., 1998) were sealed in 9 mm diameter quartz glass 213tubes, with one standard package positioned between 214 every two packages of unknowns. 215

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 cap- 220 sule positioned in the centre of the neutron field. The 221 neutron flux profile across the reactor is optimized such 222 as to give a negligible flux gradient across the central 223 12 cm of the Cd-tube. Rotation of the tube during 224 irradiation (60 min^{-1}) helps to minimize the horizontal 225 flux gradient in the tube. The correction factors for the 226 Cd-lined RODEO tube were determined in numer- 227 ous experiments in our laboratory using high purity Fe 228 doped Ca-silicate and K-silicate glass at ($^{40}\text{Ar}/^{39}\text{Ar}$)_K: 229 0.00183±0.00010, ($^{39}\text{Ar}/^{37}\text{Ar}$)_{Ca}: 0.000699±0.0000001, 230 and ($^{36}\text{Ar}/^{37}\text{Ar}$)_{Ca}: 0.000270±0.000001). 231

Upon return to the laboratory, the standard minerals 232 were loaded ca. 4-6 grains per position, (5 replicates for 233 each position) in a Cu sample tray (diameter 66 mm, 234 sample holes 2 mm diameter, 3 mm depth, 185 positions) 235 in a low volume UHV gas sample purification line 236 (Wijbrans et al., 1995) and fused by a laser single fusion 237 technique under full software control. The laser beam, 238 CW argon ion laser with principle lines at 488 nm and 239 514.5 nm and variable laser power up to 24 W in all lines 240 mode, was focused to a ca. 200 µm spot size, and under 241 software control, the x-y stage is moved in 4 circles 242 increasing in diameter from ca 500 µm to 2000 µm to 243 ensure that all individual crystals are fused using a ca. 244 15 W laser beam in the experiment. From each sample ca 245 50 mg was loaded in a Cu sample tray (diameter 66 mm, 246 22 sample holes of 6 mm diameter, 3 mm deep, and 60° 247 angle to the wall to prevent laser shadows at the bottom 248 of the pan). The rock fragments were spread out even- 249 ly in each position in the tray to ensure uniform laser 250 heating. The laser beam was defocused to a ca. 2000 µm 251 spot. The software controlled x-y stage moves the sam- 252 ple holder in a raster pattern (three runs right to left 253 direction followed by three runs perpendicular to the 254 first) under the laser beam to ensure event heating of the 255 whole sample. Laser heating under these parameters 256 lasted for 218 s, followed by 436 s clean time, which was 257 sufficient to admit clean argon gas into the mass spec- 258 trometer. The 5 isotopes of argon (m/e: 40-36) and their 259 low mass side baselines (at half mass distance) were 260 measured sequentially by magnet field controlled peak 261 hopping on an MAP 215-50 double focusing noble gas 262 mass spectrometer fitted with a Johnston MM1 SEM 263 detector operated at a relative gain of 500 with respect to 264 the Faraday collector (10¹¹ Ohm resistor on the Faraday 265 collector amplifier). The SEM amplifier is fitted with 266 three switchable resistors $(10^9, 10^8, \text{ and } 10^7 \Omega)$, that will 267 switch to an appropriate range after the ⁴⁰Ar beam in- 268 tensity is measured during the peak centering routine 269 at the beginning of each measurement. The integration 270 time for each beam is variable at 1 s increments. Typical 271

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t1.1 Table 1

Summary of ⁴⁰Ar/³⁹Ar 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 t1.2 available in electronic format

t1.3	Single fusion experiments				Incremental heating experiments								
t1.4			Age	$\pm 1\sigma$	Plateau age $\pm 1\sigma$		MSWD%39Ar, n steps Inverse Isochron $age \pm 1\sigma$				MSWD	K/Ca±1σ	
t1.5 +1.6	HAL1	VU45-A12	3297.9	± 180.4	4.08	± 0.05	1.78	36.19	3.87	± 0.17 +4.47%	1.50	0.023 ± 0.000	
t1.7 t1.8	HAL2	VU45-A14	3903.8	± 39.7	3.82	± 0.03 $\pm 0.74\%$	1.09	86.76 9	3.85	± 0.04 $\pm 0.11\%$	1.11	1.14%±0.002	
t1.9 t1.10	HAJ	VU45-A15	3803.5	±40.6	3.80	± 0.02 $\pm 0.51\%$	1.91	71.71 7	3.74	± 0.02 $\pm 0.65\%$	0.76	0.246±0.004	
t1.10 t1.11 t1.12	SzgD	VU45-A17	3959.8	±47.2	4.53	± 0.05 $\pm 1.01\%$	1.64	, 79.68 9	4.33	± 0.0376 ± 0.09 $\pm 2.11\%$	1.01	$0.120 {\pm} 0.002$	
t1.12 t1.13	SztGY	VU45-A18	4355.5	±24.6	4.22	± 0.04 $\pm 0.87\%$	1.68	86.57 9	4.14	± 0.13 +3.12%	1.60	$0.286 {\pm} 0.004$	
t1.15	HT-6	VU45-B2	7934.2	±47.4	7.94	± 0.03 $\pm 0.40\%$	1.86	46.22	7.78	± 0.07 $\pm 0.93\%$	0.37	0.075 ± 0.001	
t1.17 t1 18	HD10	VU45-B3	3671.4	±83.2	4.12	± 0.01 $\pm 0.33\%$	2.22	95.87 10	3.90	± 0.10 $\pm 2.46\%$	1.50	$0.062 {\pm} 0.001$	
t1.19	FH-4	VU45-B9	3857.9	±21.9	3.81	± 0.02 $\pm 0.49\%$	1.57	86.65	4.72	± 0.03 $\pm 0.66\%$	1.39	$0.279 {\pm} 0.004$	
t1.20 t1.21 t1.22	TW13	VU45-B11	4792.4	±25.3	4.74	± 0.02 $\pm 0.36\%$	1.91	90.51 9	4.72	$\pm 0.00\%$ ± 0.04 $\pm 0.81\%$	1.98	$0.207 {\pm} 0.003$	
t1.23	SG2	VU45-B12	5543.8	±34.1	5.48	± 0.01 $\pm 0.26\%$	1.49	54.04	5.32	$\pm 0.01\%$ ± 0.18 $\pm 3.42\%$	0.11	0.261 ± 0.004	
t1.25	KS-1	VU45-B14	4569.5	± 32.0	4.63	± 0.02 ± 0.02 $\pm 0.34\%$	2.05	71.87	4.61	± 0.02 $\pm 0.45\%$	1.92	$0.205 \!\pm\! 0.003$	
t1.27	HA-1	VU45-B15	3162.2	±23.9	3.06	± 0.02 $\pm 0.51\%$	1.17	100.00	3.01	± 0.03 $\pm 0.84\%$	0.60	$0.276 {\pm} 0.004$	
t1.29 t1.30	FT7	VU45-B17	2759.8	± 38.7	2.61	± 0.03 $\pm 1.13\%$	1.20	91.65 10	2.52	± 0.08 ± 0.08 $\pm 3.13\%$	1.12	$0.106 {\pm} 0.002$	
t1.31 t1.32	VAR	VU45-B18	4171.5	±41.7	4.08	± 0.02 $\pm 0.59\%$	0.99	81.64 5	3.85	± 0.47 +12.30%	1.23	$0.270 {\pm} 0.004$	
t1.33	AG-1	VU51-B2	2998.1	± 27.8	3.00	± 0.03 ± 0.03	1.60	99.02 9	3.14	± 0.06 +1.91%	0.98	0.100 ± 0.024	
t1.35	AG-2	VU51-B3	3692.0	± 38.8	3.30	± 0.03 ± 0.03	0.51	37.59	3.30	± 0.04 $\pm 1.110/$	0.61	$0.512 {\pm} 0.046$	
t1.30 t1.37	SP1861	VU51-B4	4153.2	± 47.8	4.15	± 0.05 $\pm 1.15\%$	0.85	98.88	3.81	± 0.18 $\pm 4.749/$	0.37	$0.025 {\pm} 0.009$	
t1.38 t1.39 t1.40	TIH	VU51-B6	7987.0	$\pm 1.15\%$ ± 28.1 $\pm 0.35\%$	7.96	± 0.03 $\pm 0.34\%$	0.51	75.89 5	8.01	± 0.07 $\pm 0.86\%$	0.46	0.164 ± 0.043	

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

Geochronology (Steiger and Jäger, 1977). Using the 285 values for flux monitors, decay constant and ⁴⁰K abun- 286 dance discussed in this paragraph in the 2–8 Ma age 287 bracket we are aware of a consistent bias of ca 1% 288 towards younger ages between our isotopic measure- 289 ments and the APTS developed for cyclically bedded 290 Neogene sediments (Hilgen et al., 1999; Gradstein et al., 291 2004; Kuiper et al., 2004, 2005). 292

4. Results

High resolution ⁴⁰Ar/³⁹Ar laser incremental heating 294 experiments were carried out on 18 samples from 14 295

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

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

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

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

5. Discussion

The new ⁴⁰Ar/³⁹Ar 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-tû (Fig. 2, Episode I). 359

355



Fig. 2. Cumulative probability diagram showing all 40 Ar/ 39 Ar age information obtained for this study. All individual step ages and their 1 σ uncertainties have been used to construct the cumulative probability curve in the diagram. Plateau ages and their 1 σ 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.

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

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.

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volcanic centres often yielded age results that were 376 indistinguishable. This observation forms the basis 377 for dividing volcanic activity of the field into 5 distinct 378 episodes (I, II, IIIa, IIIb and IV). These episodes are 379 defined as periods of activity yielding tightly clustering 380 ages, often within the individual 2 sigma uncertainties of 381 the plateau age results. Three ages, that of SG2 ($5.48\pm$ 382 0.01 Ma), AG-2 $(3.30\pm0.03$ Ma, and FT7 $(2.61\pm$ 383 0.03 Ma) have not been shown as occurring in different 384 centres. 385

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

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

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 ($\sim 260 \text{ m}$ 430 and \sim 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

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 subarieal 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 subaerial 465 magmatism in combination with a water-saturated, near 466 surface, aquifer. Conditions at this time were still sub- 467 stantially wetter then present day conditions in the area. 468

A group of volcanoes, designated as belonging to 469Episode II is slightly older then the Fekete-hegy and 470associated sites on the basis of the 40 Ar/ 39 Ar ages 471grouping 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

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

The ⁴⁰Ar/³⁹Ar ages of Szent György-hegy: Kissomlyó 505and Tóti-hegy seem to indicate an eruptive period from 5064.2 to 4.8 Ma, which overlaps in time with the period 507 when the Pannonian Lake progressively decreased in size 508(Sacchi et al., 1999). The age of the lava lake infilling the 509Kissomlyó tuff ring (Fig. 3b) is 4.63 Ma by the 40 Ar/ 39 Ar 510method. This age is significantly younger than that of the 511nearby (5.48 Ma) Ság-hegy lava, and, therefore, assuming 512coeval initiation of volcanism in the Kissomlyó-Ság-513 hegy area, it can be interpreted as the age of a lava which 514erupted from the same volcano that produced the 515 Kissomlyó tuff ring. Although all these volcanoes be-516longing to the 4.2-4.8 Ma period erupted in subaerial 517conditions, the widespread evidence of phreatomagma-518 tism is considered strong evidence for the abundance of 519 water in the rocks near the Earth's surface at this time. The 520presence of peperite, intra-crater lacustrine sediments, and 521glassy volcanic textures may reflect surface water in-522volvement in the development of the Kissomlyó volcano 523 and suggest that shallow (few metres) standing water 524bodies may have developed from time to time on the large 525flat plain of western Hungary (Martin and Németh, 2005). 526The oldest age (5.48 Ma) for the volcanic field in the 527younger age group was derived from a peperitic sill from 528Ság-hegy. This age is correlated with the last lowstand of 529the Pannonian Lake, however, the pyroclastic succession 530

and intrusive bodies of Ság-hegy clearly demonstrate

531

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

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

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

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

Several authors have suggested that there is a relation 586between magmatism and basin extension in the Panno-587nian Basin (Horváth, 1993). The main phase of basin 588 extension in the Pannonian Basin, however, predates the 589 development of the West Hungarian volcanic fields. The 590onset of magmatism at ca 7.95 Ma in fact occurred 591during a period of relative quiescence in the basin evo-592lution, and the main phase of magmatism around 4.0 Ma 593coincides with the onset of basin inversion (Cloetingh 594et al., 2005; Fodor et al., 2005). While basin inversion 595596 was probably responsible for the disappearance of the Pannonian lake in the early Pliocene, there is no clear 597 evidence that it caused the episodes of mantle melting 598recorded in the volcanic fields of western Hungary. 599During the early stages of inversion the environment was 600 601 still wet enough to cause the phreatomagmatic features in the volcanic field, but it is probably significant that one 602 of the shield volcanoes in the area, Agát-tetõ, is in fact 603 one of the youngest features in the field, and formed after 604 basin inversion had largely dried out the area. 605

606 6. Conclusion

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

larger range in expected ages and thus the increased 634 precision of the 40 Ar/ 39 Ar method also should be com- 635 plemented with more and more focused field research in 636 order to interpret the isotopic results. 637

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

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- Volcanic activity peaked around 4 million years 639 ago during perhaps 2 periods of intensified 640 activity that affected several centers. The older 641 Tihany–Hegyes-tû period at 7.95 Ma was of more 642 limited importance, both in areal extent and in 643 volume of magmatism.
- (2) Volcanism occurred near-synchonously at multi- 645 ple locations at four times during the history of the 646 volcanic field: first at ~7.95 Ma (n=2, at Tihany 647 and Hegyes-tû), at ~4.1 Ma (n=5, at Halom- 648 hegy, Szent György-hegy, Hegyesd, Vár-hegy, 649 and Sümegprága), at ~3.8 Ma (n=3, at Halom- 650 hegy, Hajagos, and Fekete-hegy), and at 3.0 Ma 651 (n=2, at Agár-tetõ, and Haláp).
- (3) There are no clear spatial patterns in the 653 distribution and timing of volcanism in western 654 Hungary, There may have been though a slight 655 east to west shift in the location of vents with 656 time.
- (4) The very low analytical uncertainties of the 658 40 Ar/ 39 Ar dates allow us to distinguish volcanic 659events at closely spaced centres to provide better 660understanding of rejuvenation of volcanic eruption 661centres at the same place (Kissomlyó vs. Ság-662hegy), and may also be used with success to 663confirm more prolonged activity at individual 664sites, and thus may cast doubt on whether this type 665of volcanism is truly 'monogenetic'. 666
- (5) The dated volcanoes erupted at a time of lowstand 667 in the nearby Pannonian Lake, and despite the 668 abundant evidence to support a water-rich erup- 669 tive environment (Ság-hegy, Kissomlyó, Tihany) 670 these volcanoes are inferred to have erupted in a 671 subaerial phreatic zone adjacent to the lake itself. 672

7.	Uncited references	673	Q1
	Bada and Horváth, 2001	674	
	Horváth and Tari, 1999	675	
	Webb et al., 2004	676	
Ac	cknowledgments	677	

This project is part of the ISES-1 scientific program of 678 the Vrije University (granted partly to JW). Partial fi- 679 nancial support from the DAAD German–Hungarian 680

Academic Exchange Program to UM and KN, the Hun-681 garian Science Foundation OTKA F 043346 granted to 682 KN, the Hungarian Science Foundation OTKAT 043344 683 granted to BK, the Magyary Zoltán Postdoctoral Fel-684 lowship and the New Zealand Science and Technology 685 Postdoctoral Fellowship grants to KN are acknowl-686 edged. Review of an earlier version of the manuscript by 687 Tibor Dunai (University of Edinburgh) is also acknowl-688 edged. The project has benefited from discussion 689 with Gábor Csillag and Tamás Budai both from the 690 Geological Institute of Hungary. Constructive reviews 691 by James D.L. White (Otago University, Dunedin) and 692 693 an anonymous referee helped to clarify many aspects of this paper, many thanks for them. 694

695 Appendix A. Supplementary data

Supplementary data associated with this article
can be found, in the online version, at doi:10.1016/j.
jvolgeores.2007.05.009.

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