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P. Árkai · K. Balogh · I. Dunkl

Timing of low-temperature metamorphism and cooling of the Paleozoic and Mesozoic formations of the Bükkium, innermost Western Carpathians, Hungary

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Abstract K-Ar ages of illite-muscovite and fission track ages of zircon and apatite were determined from various lithotypes of the Bükkium, which forms the innermost segment of the Western Carpathians. The stratigraphic ages of these Dinaric type formations cover a wide range from the Late Ordovician up to the Late Jurassic. The grade of the orogenic dynamo-thermal metamorphism varies from the late diagenetic zone through the 'anchizone' up to the 'epizone' (chlorite, maximally biotite isograd of the greenschist facies). The K-Ar system of the illite-muscovite in the $<2\ \mu m$ grain-size fraction approached equilibrium only in 'epizonal' and high-temperature 'anchizonal' conditions. The orogenic metamorphism culminated between the eo-Hellenic (160–120 Ma) phase connected to the beginning of the subduction in the Dinarides, and the Austrian (100–95 Ma) phase characterized by compressional crustal thickening. No isotope geochronological evidence was found for proving any Hercynian recrystallization. The stability field of fission tracks in zircon was approached using the thermal histories of the different tectonic units. A temperature less than 250° C and effective heating time of 20–30 Ma had only negligible effects on the tracks, whereas total annealing was reached between 250 and 300° C. Apatite fission track ages from the Paleozoic and Mesozoic formations show that the uplift of the Bükk Mountains occurred only in the Tertiary (not earlier than ca. 40 Ma ago). Thermal modeling based on apatite fission track length spectra and preserved Paleogene sediment thickness data proved that the Late Neogene burial of the recently exhumed plateau of the Bükk Mountains exceeded 1 km.

Key words Low-temperature metamorphism · K-Ar dating · Fission track dating · Illite 'crystallinity' · Carpathians

Introduction

Radioisotopic methods have been widely used for age determination of very low-grade metamorphic rocks during the last two decades (for a review, see Hunziker 1987). The determination of closure temperatures of different isotope systems in various minerals proved to be of great importance (Dodson 1973; Purdy and Jäger 1976; Wagner et al. 1977; Harrison and McDougall 1982). In addition to the temperature, which is regarded as the decisive factor, the duration of heat effect and the heating and cooling rates considerably influence the kinetics of isotopic exchange reactions. The geochronological interpretation of the radioisotopic data from low-temperature metamorphic terranes is hindered by the following factors

1. Small grain size may considerably influence the capture of radiogenic daughter nuclides and makes the separation of pure monomineralic fractions practically impossible
2. Grain-size fractions often contain mixtures of newly formed and detrital minerals. The closure temperature range of a given mineral also varies as a function of grain size and structural state
3. The temperature range of very low-grade metamorphism overlaps the closure range of K-Ar system in illite-muscovite. Consequently, special attention should be paid to the distinction between 'metamorphic' ages, which refer to the time of crystallization at or near to the metamorphic climax, and 'cooling' ages, which indicate the time when the K-Ar system became closed.

According to Hunziker (1987), all of these disturbing effects are related to the low diffusion and reaction rates and, consequently, to the frequent non-equili-

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Péter Árkai (✉) · István Dunkl
Laboratory for Geochemical Research, Hungarian Academy of Sciences, H-1112 Budapest, Budaörsi út 45, Hungary.
Tel./Fax.: 361 1851781

Kadosa Balogh
Institute of Nuclear Research, Hungarian Academy of Sciences, H-4001 Debrecen, Bem tér 18/c, Hungary

brium state of the very low-grade metamorphic mineral assemblages.

The geochronological interpretation of the K-Ar data presented here is based largely on the results of Hunziker et al. (1986). According to their observations, the K-Ar isotopic system of illite-muscovite in the <2 μm grain-size fraction was completely reset at $260 \pm 20°$ C over 10 ± 5 Ma. In prograde diagenetic-metamorphic series continuous rejuvenation of K-Ar ages was observed with increasing temperature. This phenomenon is characterized by the decreasing amounts of smectitic mixed layers in illite at lower temperatures, and by the decrease in interlayer H_2O and the increase in the interlayer K content and the $2M_1/1M_d$ ratio. All of these processes are reflected in the decrease in illite 'crystallinity'. Complete resetting of the smaller grains was observed at lower temperatures than for larger grains (Hunziker 1987), corresponding to an exponential relation between the grain size and the parameters of diffusion as described by Fick's second law. In case of illite-muscovite with grain sizes of 20–60 μm, this temperature is about 350° C, which agrees with the closure temperature of the K-Ar system of muscovite given by Purdy and Jäger (1976).

Extrapolating from the annealing experiments which resulted in the 'healing' of fission tracks, and comparing these results with the field observations, effective closure temperatures of 100 and 200–250° C were applied to apatite and zircon (see Hurford in Hunziker 1987). Considering the kinetics of the process, a closure interval of 70–125° C/10^6–10^8 years can be used for apatite (Gleadow et al. 1983).

The aims of the present paper are: (i) to determine the lithology controlled effects of detrital white mica being present in the <2 μm fractions on the K-Ar dates as a function of metamorphic grade; (ii) to estimate the strongly debated stability and annealing fields of fission tracks in zircon by comparing the illite-muscovite K-Ar ages, zircon fission track ages and illite 'crystallinity' data; and (iii) to outline the time-temperature (tT) relation of regional metamorphism by integrating the isotope geochronological data with the petrological results for the example of the Paleozoic and Mesozoic formations of the Bükkium.

Geological setting

The Paleozoic and Mesozoic sequences of the Bükkium formed in the north-western part of the Inner Dinarides, and occupied their present position, i.e. the innermost part of the Western Carpathians, by large-scale meso-Alpine horizontal displacements of plate fragments or microplates (Fig. 1; see Balogh 1964; Kovács 1982, 1989a; Kázmér and Kovács 1985, 1989; Balla 1988). The Bükkium (or Bükk Unit) consists of the Szendrő, Uppony and Bükk Mountains. The main geological, structural and metamorphic petrological features are summarized in Fig. 2 (for details of metamorphism, see Árkai 1983, 1991).

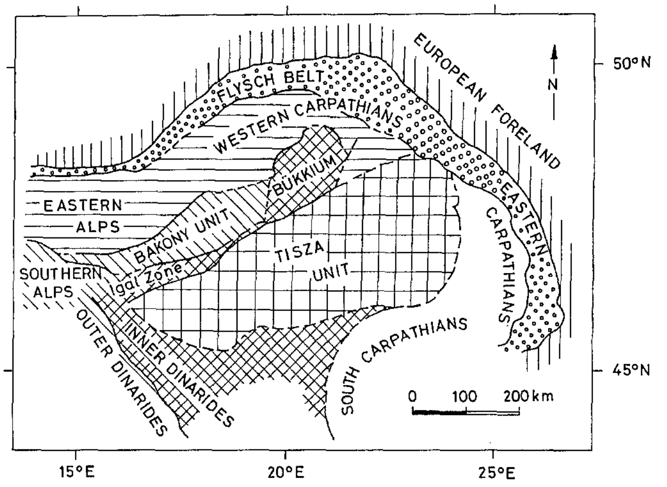

Fig. 1 Tectonic setting of the Bükkium within the Alp-Carpathian-Dinaric framework

The Paleozoic (Middle Devonian to Middle Carboniferous) formations of the Szendrő Mountains, (unit 2 in Fig. 2) underwent transitional low- to intermediate-pressure, 'epizonal' (greenschist facies, chlorite zone) regional metamorphism. Locally, in the southern part of the area, the biotite isograd was reached. The fine, dispersed coalified matter is in the graphite-d_1 state (classification after Landis 1971). A metamorphic temperature range of 350–450° C is assumed, also taking into consideration the carbonate (calcite-dolomite) thermometric data of Árkai and Fórizs (unpublished data) and the temperature estimates of the biotite isograd of Winkler (1979) and Ferry (1984).

In the Uppony Mountains the Upper Ordovician to Middle Carboniferous formations (unit 3 in Fig. 2) suffered low-pressure regional metamorphism with temperature corresponding to the boundary between the 'anchi-' and 'epizones' (ca. 350° C). In metapelites of the Uppony Paleozoic, coexistence of graphite-d_2, meta-anthracite and metabituminite was found. Metasandstones contain chloritoid; metabasaltic lavas and tuffs contain assemblages spanning the glauconite-celadonite/stilpnomelane isograd.

The Bükk Mountains consisting of Middle Carboniferous to Late Jurassic formations, are built up by several tectonic units, in which the stratigraphic sequences and metamorphic conditions vary (Csontos 1988; see also Fig. 2). The Fennsík (Bükk Plateau) Parautochton unit of the Bükk Mountains (units 5–7 in Fig. 2) suffered mostly anchizonal, partly epizonal, low- to medium-pressure regional metamorphism. The Szarvaskő–Mónosbél Nappe, i.e. the uppermost tectonic unit of the Bükk Mountains (9 in Fig. 2) is formed of Jurassic clastic sequences which suffered late diagenetic and low-temperature anchizonal transformations. The secondary mineral associations of the MORB-type ultra-

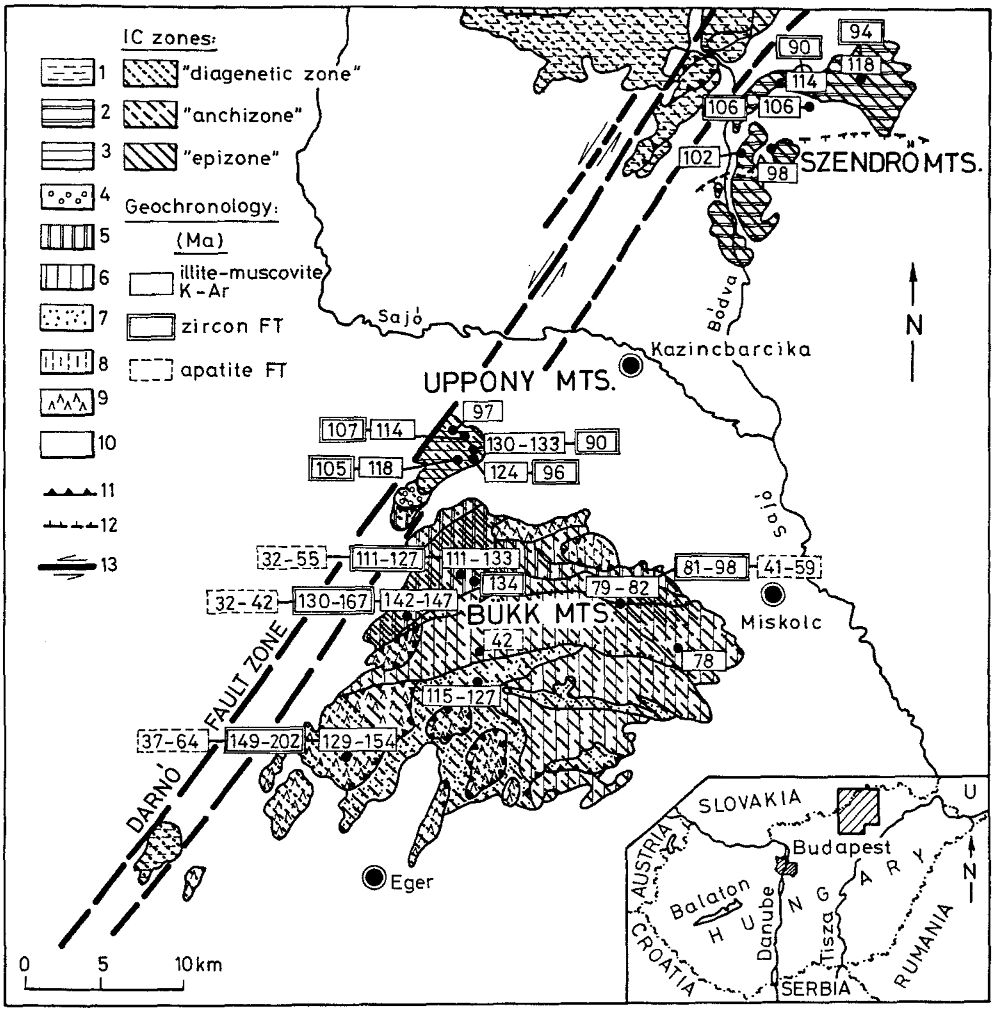

Fig. 2 Geological and metamorphic sketch map of the Bükkium after Kovács (1989b), Csontos (1988) and Árkai (1983, 1991), also showing the geochronological data. Legend: 1, South Gemer Unit (Upper Permian-Mesozoic); *2,* Szendrő Paleozoic (Middle Devonian-Middle Carboniferous); *3,* Uppony Paleozoic (Upper Ordovician(?)-Middle Carboniferous); 4, Gosau-type Senonian conglomerate; 5–9, Bükk Mountains Paleo-Mesozoic: 5–7, Fennsík (Bükk Plateau) Parautochthon: 5, Paleozoic (Middle Carboniferous-Permian); 6, Triassic (Bükk Plateau and Eastern Bükk); 7, Jurassic (Southern Bükk); 8, Kisfennsík (Little Plateau) Nappe (Triassic); 9, Szarvaskő-Mónosbél Nappe (mostly Jurassic); 10, Tertiary and Quaternary; 11, nappe boundary; 12, major imbrications within the nappes; 13, strike-slip fault. In the case of dates, single values or intervals ($n > 5$) are given

basic-basic-acidic complex at Szarvaskő (Western Bükk) correspond to the prehnite-pumpellyite facies.

Based on the available metamorphic petrological data, a single-phase, prograde metamorphic event was proved for the Paleozoic and Mesozoic formations of the Bükkium. The age of metamorphism is Alpine (Cretaceous, pre-Senonian), as deduced from stratigraphic, tectonic and petrological evidence (Árkai 1973, 1983). No signs of pre-Alpine orogenic metamorphism could be proved.

Before this study only sporadic isotopic age data were obtained on the Bükkium. Using whole-rock samples Alpine (Austrian, 93–102 Ma) effects were documented on the Upper Triassic metarhyolite of the Eastern Bükk Mountains by K-Ar and Rb–Sr methods (Balogh et al. 1980; Kovách et al. 1985). The age of the basic-ultrabasic magmatism in the Western Bükk (at the village of Szarvaskő) is 165±5 Ma as determined by K-Ar data on magmatic amphibole and contact metamorphic muscovite (Árva-Sós et al. 1987). Whole-rock K-Ar data of the metabasalts from the Uppony Mountains (village of Nekézseny) refer partly to the Cretaceous post-magmatic effect, and can partly be considered as mixed ages (110–157 Ma, Balogh et al. 1989).

Methods

Isotope geochronological measurements were preceded by petrographic [mesoscopic, microscopic and X-ray powder diffractometric (XRD)] investigations to select the appropriate representative samples from a large population of more than 1000 samples, which formed the base of the metamorphic petrogenetic reconstruction given by Árkai (1983). The procedure of sample preparation and the petrographic techniques used were described earlier (Árkai 1983, 1991). To assure the possibility of inter-laboratory correlation of illite 'crystallinity' (IC) data and scales, only some details of XRD work are given here.

Illite 'crystallinity' (IC) was measured after Kübler (1967, 1975, 1990). The calibration of IC values against those of Kübler's laboratory (where the 0.25–0.42 $\Delta°2\Theta$ boundary values of the anchizone were established) was performed using the standard rock slab series Nos 32, 34 and 35 provided by B. Kübler. Thus in the present study the boundary values of Kübler's anchizone correspond to 0.28–0.44 $\Delta°2\Theta$, measured on sedimented, air-dried mounts of the <2 μm grain-size fractions. However, the 'diagenetic', 'anchi-' and 'epizone' were re-defined by Árkai (1983, 1991) using the correlations between IC, clay mineral assemblages, vitrinite reflectance and metabasite mineral facies. The IC boundaries of this 'anchizone' are 0.25–0.34 $\Delta°2\Theta$ on the scale of the present work, and correspond to 0.21–0.31 $\Delta°2\Theta$ on Kübler's original scale. This 'anchizone' is correlated roughly with the pumpellyite-actinolite facies and with the medium and high-temperature parts of the prehnite-pumpellyite facies, and is characterized by vitrinite reflectance ranges of R (random) ≈ 5.0–3.0% and R (max) ≥ 6.0–3.5%. The estimated temperature range of Kübler's anchizone is 200–300° C, whereas that of the re-defined 'anchizone' corresponds to 250–350° C (see Frey 1986; Kisch 1987; Árkai 1991). These re-defined zones (indicated by quotation marks) are used in the present paper. For further details about the theoretical and methodological problems of the determination of zone boundaries, see Árkai (1991) and Kisch (1990).

K-Ar dating was performed with instruments constructed in the Institute of Nuclear Research, Hungarian Academy of Sciences. Samples were degassed by high frequency induction heating; argon was cleaned using the usual method of applying zeolite, cold traps and furnaces with a Ti sponge and CuO. ^{38}Ar was introduced with a gas pipette. For Ar isotopic ratio measurements a magnetic mass spectrometer of 150 mm radius and 90° deflection was used in the static mode. Before K determination the samples were digested by a mixture of $HF + H_2SO_4 + HClO_4$ and dissolved thereafter in HCl. Sodium buffer and Li internal standards were added and the K content was measured with a flame emission photometer. The inter-laboratory standards Asia 1/65 and GL-O were used for controlling and the calibration of Ar and K determinations. Errors of K-Ar ages (2σ) were calculated assuming a 3% error for the determination of K. This includes a 1.5% estimated error for the standards Asia-1/65 and GL-O and a 1% error for the Ar isotope ratios. The latter is related mainly to the possible fractionation in the Ar extraction line and to a certain background peak at mass 36. The results obtained on inter-laboratory standards have also been published by Odin et al. (1982).

To avoid the disturbing effects of detrital muscovite, the K-Ar ages obtained on the <2 μm (and, in certain cases, on the 2–0.6 and <0.6 μm) fractions were used for dating the metamorphism, following the practice of Clauer and Kröner (1979), Frank and Stettler (1979), Bonhomme et al. (1980), Hunziker (1979, 1987), Hunziker et al. (1986) and Reuter (1987). It is generally assumed that these small grain-size fractions are mostly devoid of detrital mica, although the minor effects of detrital muscovite can be demonstrated even at high-temperature anchizonal conditions (Árkai 1983; Reuter 1987). The effect of detritus on K-Ar ages of metaclastic rocks can also be checked by investigating the intercalated metatuffs, in which white K-mica is of exclusively metamorphic origin (see Ahrendt et al. 1978). The impurities (e.g. quartz, albite, chlorite, rutile) usually present in the commonly used <2 μm fractions do not significantly affect the reliability of the K-Ar ages (Hunziker et al. 1986).

For fission track dating zircon and apatite grains were separated from the crushed and sieved 70–180 μm fraction by bromoform and a magnetic separator; they were then hand-picked under a binocular microscope. The apatite crystals were embedded in epoxy resin, the zircons in FEP-Teflon. For apatite, 1% nitric acid was used with a 2.5–3 min etching time (Burchart 1972). In the case of zircon crystals, an eutectic melt of NaOH-KOH-LiOH was used at lower temperatures (190° C) than suggested by Zaun and Wagner (1985). Neutron irradiations were made at the nuclear reactor of the Technical University, Budapest. The external detector method was used for both minerals (Gleadow 1981). After irradiation, the induced fission tracks in the mica detectors were etched by 40% HF for 40 min. Spontaneous track counts were made in oil immersion under a Zeiss NU 2 microscope, with a magnification of ×1600; for mica, external detectors with dry optics of ×800 magnification were used.

The fission track ages were determined by the zeta method (Hurford and Green 1983) using zircon age standards from the Fish Canyon Tuff, Buluk Member Tuff and Tardree Rhyolite and apatite standards from Durango and Fish Canyon Tuff. Reference ages of 27.8 ± 0.2 Ma for the Fish Canyon Tuff, 31.4 ± 0.5 Ma for the Durango apatite, 16.2 ± 0.6 Ma for the Buluk Member Tuff and 58.7 ± 1.1 Ma for the Tardree Rhyolite have been adopted according to Hurford and Hammerschmidt (1985), Green (1985), Hurford and Watkins (1987) and Hurford and Green (1983). The error was calculated using the classical procedure, i.e. by the double Poisson dispersion (Green 1981).

The chlorine content of the apatites was determined by electron microprobe to estimate whether the grains could be used for thermal modeling. The analyses were carried out on a JEOL JCXA–733 electron microprobe equipped with three wavelength-dispersive X-ray spectrometers.

For geological interpretation of the isotopic age data, the time-scale of Harland et al. (1989) was used.

Chronology of metamorphism

Localities, rock types, stratigraphic ages, apatite and zircon fission track and illite-muscovite K-Ar ages and IC values of the samples characteristic of the different tectonic units of the Bükkium are listed in Table 1. The analytical and statistical results of the K-Ar isotopic and fission track measurements can be obtained from the first author on request.

In Fig. 3 the radioisotopic ages are plotted as a function of IC. With increasing IC (i.e. with decreasing metamorphic grade or temperature) the differences between the K-Ar and zircon fission track values increase. This discrepancy relates to the disturbing effects of detrital phases. An acceptable overlap between the two

Table 1 Rock types, stratigraphic, fission track and K–Ar ages and illite 'crystallinity' (IC) values of the samples representing the main tectonic units of the Bükkium*

Sample	Rock type	Stratigraphy	K–Ar ages (Ma)†			Fission track ages (Ma)		IC (Δ°2Θ)
			<2 μm	0.6–2 μm	<0.6 μm	Zircon	Apatite	
Bükk Mountains, Szarvaskő-Mónosbél Nappe (unit 9 in Fig. 2)								
90-B-1	Sandstone	J	129±5			178±20	49±29	0.362
90-B-2	Conglomerate	J				176±22	47±7	
90-B-3	Sandstone/shale	J				153±17	65±7	
90-B-5	Sandstone	J				154±20	56±9	
90-B-6	Sandstone	J	152±6					0.488
90-B-7	Sandstone	J				149±24 164±20	64±8	
90-B-8	Sandstone	J	137±5			196±25	59±6	0.371
90-B-20	Sandstone olistolith	J				184±22 202±28	37±8	
B-146/a-b	Sandstone/shale	J	154±6	152±6	127±5		55±5	0.446
Sz-44	Gabbro pegmatite	J					50±9	
Sz-43	Gabbro	J					57±12	
Sz-46	Gabbro	J					48±26	
Sz-47	Gabbro	J					53±9	
Bükk Mountains, Fennsík (Bükk Plateau) Parautochthon (units 5–7 in Fig. 2)								
Jurassic slate formation, Southern Bükk (unit 7)								
Lökvölgy-II/zs	Slate	J	115±4	120±4	102±4			0.281
Lökvölgy-II	Slate	J	119±5	113±4	86±3			0.288
90-B-4	Sandstone	J	127±5					0.362
Triassic formations of the Eastern Bükk (unit 6)								
90-B-11	Psammitic slate	T_3	79±3			82±8 98±10	43±7	0.216
90-B-13	Diorite	T					41±7 46±13	
B-600	Meta-andesite tuff	T_2		82±3				0.207
90-B-10	Metabasalt tuff	T_3					59±10 41±16	
Bagolyhegy-2	Metarhyolite tuff	T_3		77±3	67±3			0.183
Bagolyhegy-3	Metarhyolite tuff	T_3		78±3	45±2			0.174
Carboniferous and Permian of the Northern Bükk (unit 5)								
90-B-14	Metasandstone	P_1				133±23 135±16		
Tr-II/8	Slate	C_2	133±5	133±5				
90-B-15	Metasandstone/slate	C_2				124±16	32±6	
Tr-II/b	Metasandstone	C_2				127±16	53±6	0.239
90-B-16	Metasandstone	C_2	111±4			111±12	55±7	0.214
B-170	Slate	C_2	122±5					0.219
90-B-17	Conglomerate/slate	C_2	147±6			176±20	42±8 39±11	0.222
90-B-18	Congl./metasandstone	C_2	142±5			130±16	32±6	0.231
Uppony Mountains (unit 3)								
U-1945 m	Metagreywacke	C_{2-3}	114±4			107±14		0.234
U-380 m	Cipollino	D_3	97±4					0.198
Dt-8. 31.0 m	Slate	S(?)	130±5	133±6				0.177
Dt-8. 268.0 m	Slate	S(?)	133±5	133±5				0.221
Dt-8. 276-8 m	Metasandstone	S(?)				90±11		
U-7012	Metasandstone	S(?)	118±5			105±13		0.181
U-7010	Metasandstone	O_3(?)		123±5	71±3	98±17		0.211
U-7002	Metasandstone	O_3(?)		125±5	90±4	94±12		0.276
Szendrő Mountains (unit 2)								
Sz-22. 33.8 m	Phyllite	C_{1-2}		102±4				0.200
G-1. 352.3 m	Phyllite/metasandstone	C_{1-2}	106±4			106±15		0.200
Rsz-5. 35.0 m	Metasandstone	C_{1-2}	114±5			90±14		0.211
M-3. 9.0 m	Metasandstone	C_{1-2}	118±5			94±11		0.249
A-1. 61.3 m	Cipollino	D_3		98±4				0.224

* The analytical and statistical data of age determinations and a description of the samples, including localities (with map), microstructural features and modal compositions, can be obtained from the first author on request.

† Calculated with atomic constants suggested by Steiger and Jäger (1977).

Legend: J, Jurassic; T, Triassic; P, Permian; C, Carboniferous; D, Devonian; S, Silurian; and O, Ordovician.

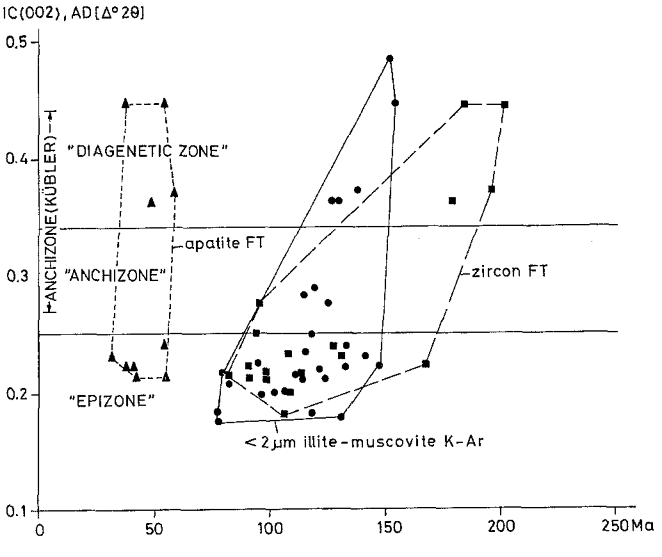

Fig. 3 Plot of geochronological data versus illite 'crystallinity' (IC)

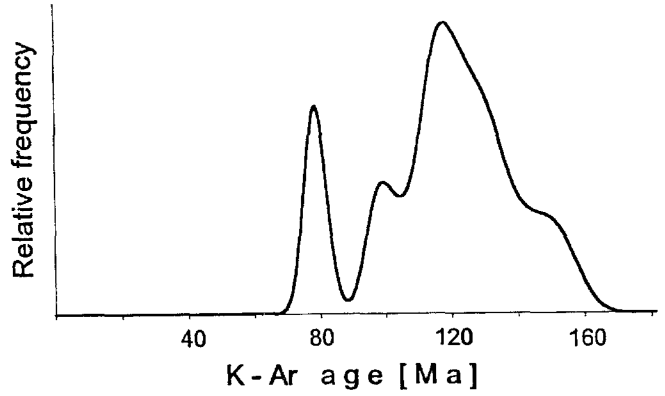

Fig. 4 'Age spectrum' of all K-Ar ages of the Bükkium, compiled by the method of Hurford et al. (1984). The samples from the eastern part of the Parautochthon form the youngest, sharp peak of ages with an average of 79±3 Ma. The main population (with a peak of 118 Ma) contains all the other samples. The right-side asymmetry and the 'shoulder' at the older side of the peak (ca. 150 Ma) are related to the partly or unmodified detrital ages

data sets can be observed only in 'epizonal' and high-temperature 'anchizonal' environments. Apatite ages are practically independent of the metamorphic conditions and refer to a later stage of thermal evolution (cooling). Figure 2 demonstrates the regional distributions of the different isotopic age data.

In the 'epizonal' metamorphic Paleozoic formations of the Szendrő Mountains (unit 2 in Fig. 2) the illite-muscovite K-Ar average (108±8 Ma) and the zircon fission track average (97±8 Ma) are interpreted as cooling ages. The highest K-Ar age (118 Ma) on <2 µm illite-muscovite was obtained from a metasandstone, whereas the lowest value (98 Ma) was obtained from a cipollino-like marble which contains metatuffitic intercalations.

In the transitional 'anchi-/epizonal' Paleozoic of the Uppony Mountains (unit 3 in Fig. 2) the average of the illite-muscovite K-Ar dates is 118±14 Ma; that of the zircon fission track dates is 99±7 Ma. (In case of the 'epizonal' Szendrő Paleozoic, a similar trend, but smaller difference, was found, which proved to be not significant even at $P=10\%$). The highest K-Ar age (133 Ma) was found in a slate, whereas the youngest (97 Ma) was found in a cipollino-like metatuffitic limestone. (The latter difference can be explained by the slight effect of detrital muscovite in slates. This indicates that the temperature of the 'anchi-/epizonal' transition might be insufficient to reach the total resetting of the Ar system in the <2 µm illite-muscovite.)

Not only the metamorphic grade and pressure indicating parameters, but also the isotopic ages, vary between the main tectonic units of the Bükk Mountains.

The K-Ar age of the high-temperature 'anchizonal' Jurassic slate of the Fennsík (Bükk Plateau) Parautochthon in the Southern Bükk (unit 7 in Fig. 3) measured on the <2 µm illite-muscovite varies between 115 and 119 Ma, which agrees fairly well with the illite-muscovite K-Ar ages from the Paleozoic formations of the Szendrő and Uppony Mountains. The K-Ar data of the <0.6 µm grain-size fraction of the Jurassic slate (86–102 Ma) correspond to the K-Ar ages of the metatuffitic rock types of the Szendrő and Uppony Mountains (in these instances the effects of the detrital micas might be minimal or absent).

Significantly younger age data were obtained in the Triassic formations of the eastern part of the Parautochthon (unit 6 in Fig. 2). Here the K-Ar and the zircon fission track ages of the metaclastic rocks (79 and 82–98 Ma, respectively) are in agreement with the K-Ar ages of the meta-igneous rocks (77–82 Ma). The sharp deviation of K-Ar results of the eastern part of the Parautochthon from the other K-Ar ages of the Bükkium is shown in Fig. 4. This discrepancy can be interpreted either by regional differences in the cooling histories of the various parts of the Parautochthon or by a Late Cretaceous ductile deformation along a NW-SE trending shear zone, the importance of which has been pointed out by Csontos (personal communication).

The northern and the north-western part of the Parautochthon comprises Carboniferous and Permian metasedimentary rocks (unit 5 in Fig. 2). Despite the relatively strong, commonly 'anchizonal', partly 'epizonal' metamorphism, both the K-Ar and the zircon fission track dates suggest non-equilibrium conditions of recrystallization. The scatter of the data is large: the K-Ar dates vary between 111 and 147 Ma, whereas the zircon fission track dates are between 111 and 167 Ma, which may refer to the effects of detritus in both systems. In this instance the youngest age value may give the maximum age of metamorphism.

In the Szarvaskő–Mónosbél Nappe of the Bükk Mountains (unit 9 in Fig. 3) the heat effect of the low-temperature (commonly 200° C, maximally 250–300° C) 'diagenetic'-'anchizonal' alteration was not sufficient to

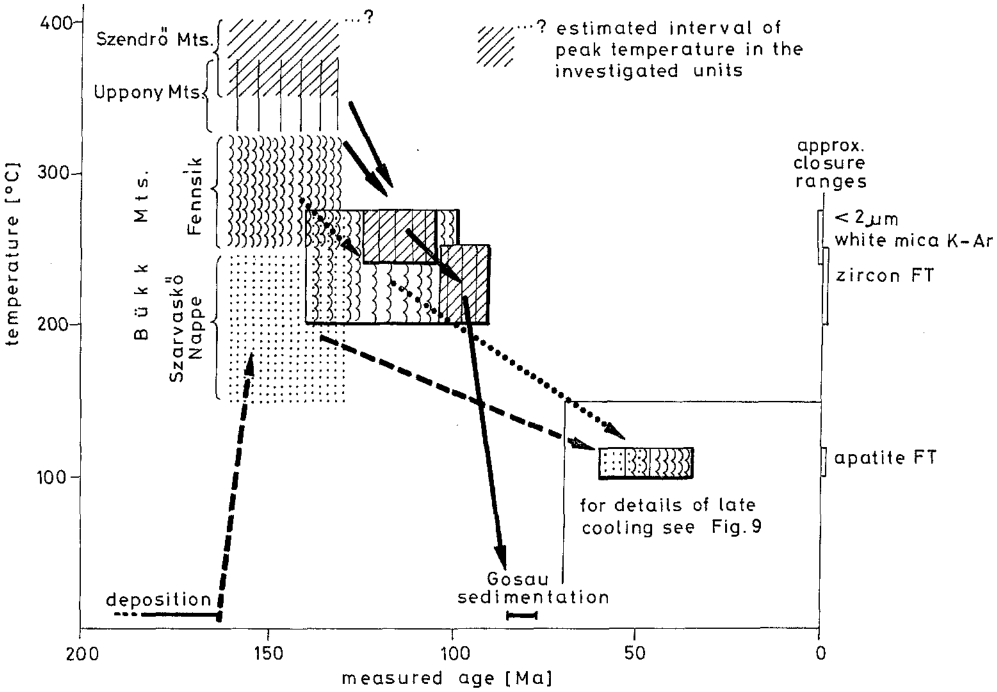

Fig. 5 Time-temperature (*tT*) paths of the main tectonic units of the Bükkium. FT, fission track

reset the zircon fission track clock: the data scatter between 149 and 202 Ma. These data are considered as mixed ages: the effects of the pre- or syn-Jurassic detritus and the Cretaceous metamorphism were superimposed. The differences between the zircon fission track and illite-muscovite K-Ar data are the largest in case of these lowest grade formations. The K-Ar data are also strongly scattering between 129 and 154 Ma, indicating the effect of detrital muscovite in the <2 μm grain-size fraction.

Figure 5 summarizes the *tT* evolution paths of the main tectonic units of the Bükkium. The difference in late cooling histories of the Szendrő and Uppony Paleozoic on the one part and of the Bükk Mountains, on the other, as displayed in Fig. 5 is also evidenced by the stratigraphic and lithological observations of Brezsnyánszky and Haas (1984) and Balogh (1964). Further details are given in a later section.

Long-term natural annealing of fission tracks in zircon

The thermal stability of fission tracks in zircon is strongly debated. For solving this question two different approaches have been used (1) extrapolation from laboratory annealing experiments and (2) multi-method dating of slowly cooling igneous and metamorphic terranes. These approaches resulted in a broad spread of closure temperatures between 175 and 340° C (Harrison et al. 1979; Lal et al. 1980; Nagpaul 1982; Bal et al. 1983; Hurford 1986). The contradictory feature of the experimental results (displayed in Fig. 6) may be related to the differences in chemistry and metamictization of the grains and to the diverse etching conditions.

In contrast with apatite, our knowledge of the natural annealing of zircon is incomplete. The works of Harrison et al. (1979), Hurford (1986) and Zaun and Wagner (1985) were related to uplifting terranes. Although their calculations of the closure temperature are well established for different cooling rates, the time-temperature dependence is not clear, because these formations presumably cooled monotonously from high-grade metamorphic conditions. Fission track dating of metasedimentary and/or metavolcanic formations with well-known burial metamorphic *tT* paths would be the only way to this problem.

In the case of Bükkium, the time and temperature limits of the low-temperature metamorphism are fairly well known. The beginning of heating is controlled by the termination of sedimentation not earlier than Oxfordian times. The upper time limit is given by the post-metamorphic cooling ages (obtained on units of higher metamorphic grade). This time span is about 35–40 Ma, thus the duration of effective heating could not be greater than 20–30 Ma. In Fig. 6 the rectangles represent the time-temperature fields of metamorphism. The increase in the rejuvenation with metamorphic temperature is evident. It can be concluded that the temperature of the total annealing of such a heating period might be between 250 and 300° C. The beginning of the track annealing is more doubtful (similar to apatite), but it is remarkable that metamorphic temperatures ranging up to 250° C did not produce total annealing (see the zircon fission track dates of the Szarvaskő-Mónosbél Nappe of the Bükk Mountains: unit 9 in Fig. 2).

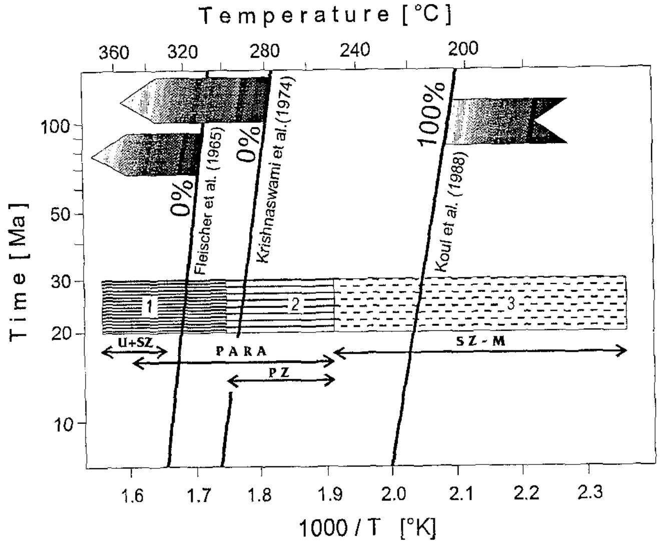

Fig. 6 Arrhenius plot of the annealing of fission tracks in zircon and the trend observed in the Bükkium. The values of 0 and 100% refer to the maximum temperature of no track annealing and the complete erasure of tracks, respectively. The absolute disagreement between the results is clearly shown: according to Koul et al. (1988) no tracks are stable to the left of the line around 200° C, whereas according to the experiments of Krishnaswami et al. (1974) and Fleischer et al. (1965) there is no annealing to the right of lines between ca. 280 and 320° C. Rectangles indicate the estimated effective heating time and temperature ranges of the different tectonic subunits of the Bükkium. Legend: U+SZ, Paleozoic formations of the Uppony and Szendrő Mountains; PARA, Fennsík Parautochthon of the Bükk Mountains; PZ, Paleozoic of the Parautochthon of the Bükk Mountains; SZ-M, Szarvaskő-Mónosbél Nappe; 1, total rejuvenation of the zircon fission track ages; 2, nearly complete rejuvenation; and 3, negligible changes

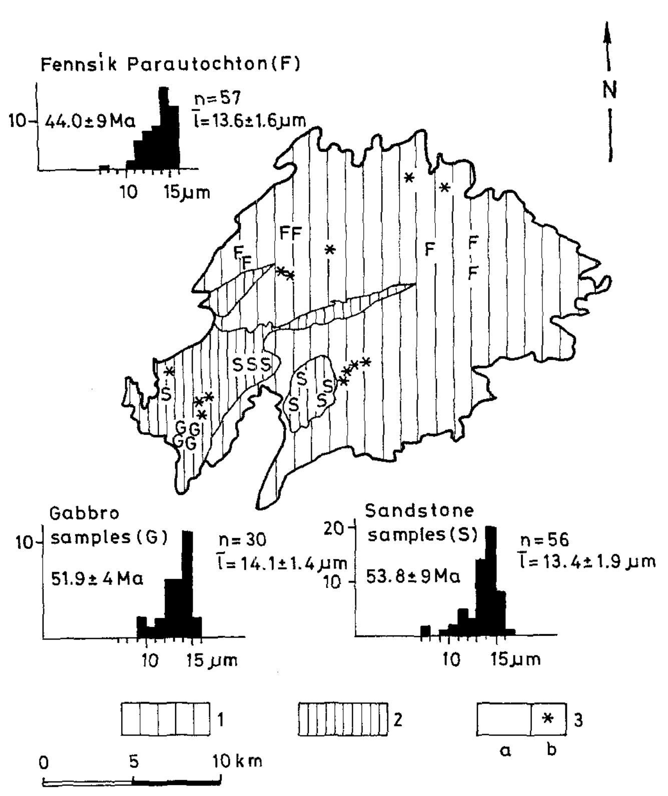

Fig. 7 Apatite fission track length distributions from the Bükk Mountains. Legend: 1, Fennsík Parautochthon (Late Paleozoic-Mesozoic): F, different lithotypes from the Fennsík Parautochthon; 2, Szarvaskő-Mónosbél Nappe (mostly Jurassic); G, gabbro; S, metasandstone; 3, Neogene cover (a) basin-filling around the mountains and (b) smaller remnants preserved at high altitudes

Post-metamorphic cooling and uplift history of the Bükk Mountains

The discussion about the late cooling is restricted to the tectonic units of the Bükk Mountains, as no apatite fission track ages could be obtained from the Paleozoic of the Szendrő and Uppony Montains. The apatite fission track data show that these units cooled below the closure temperature range of 125–70° C during the Tertiary, between the Paleocene and the Oligocene. The uppermost tectonic unit (the Szarvaskő-Mónosbél Nappe) reached this temperature interval earlier; its apatite fission track ages are older (on average 53 ± 8 Ma) than those of the underlying Fennsík Parautochthon (on average 43 ± 8 Ma).

Because of the rather complex structural/burial history of the Bükk Mountains, the apatite fission track results can only be considered as apparent ages. Apatites show negatively skewed confined track length distributions with no very short (<9 μm) tracks; the means range from 13.6 to 14.1 μm with standard deviations of 1.4 to 2 μm (Fig. 7). The main populations of the spectra display a mild shortening compared with the length distributions of thermally undisturbed volcanic formations. The shapes of the length distributions of the intrusive bodies and the metasandstones of the Szarvaskő-Mónosbél Nappe are similar; however, the length of the main groups is larger in the former.

Green et al. (1986) showed that the increasing Cl^- substitution in the lattice of apatite increases the thermal stability of the tracks. The thermal modeling procedures were mainly based on the annealing experiments of the Durango apatite with a Cl/F ratio of about 1:8 (Green et al. 1986). Figure 8 displays the Cl versus F contents of the apatites from the dated rocks of the Bükk Mountains. Apatites from the various lithotypes show differences in Cl/F ratios. However, most of the results cluster around the composition of the Durango standard. Consequently, the thermal modeling procedure is applicable to the investigated samples.

The set of geological/burial input data for thermal modelling is imperfect due to recent uplift and advanced erosion. However, the following geological observations were used to place limits on the time of exhumation of the Paleozoic and Mesozoic units, and thus to constrain the thermal history.

The Gosau-type coarse clastic rocks of Santonian-Campanian age unconformably overlying the Paleozoic of the Uppony Mountains represent the first record of post-metamorphic sedimentation in the Bükkium.

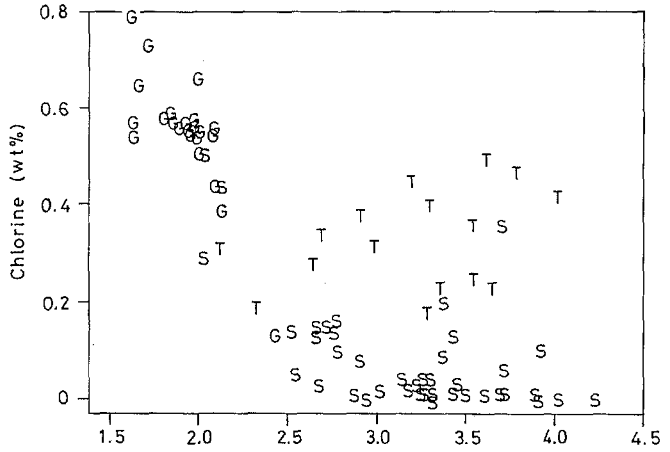

Fig. 8 Distributions of chlorine and fluorine contents of apatite from different lithotypes. Key: G, gabbro and S, metasandstone from the Szarvaskő-Mónosbél Nappe; T, Triassic tuffaceous slate from the Fennsík Parautochthon

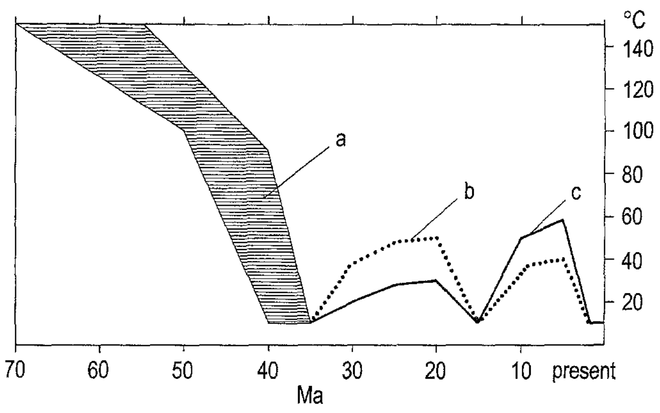

Fig. 9 Possible post-Cretaceous cooling paths of the Bükk Mountains based on thermal modeling of the fission track data. Key: (a) envelope of the acceptable pre-Priabonian cooling paths; (b) considering the maximum thickness of the Paleogene sediments; and (c) considering negligible Paleogene thickness and maximal post-Karpatian burial

These non-metamorphic rocks do not contain pebbles deriving from the Bükk Mountains (Brezsnyánszky and Haas 1984).

In the Bükk Mountains a red terrestrial sediment of unknown age is the oldest cover on the Paleo-Mesozoic formations. The marine sedimentation began in the Late Eocene (Priabonian, see Balogh 1964). The thickness of the Oligocene–Lower Miocene beds in the neighborhood of the Bükk Mountains is variable, generally 500–1000 m, but in the basins exceeds 1500–2000 m (Csiky 1961; Báldi 1986).

West of the Bükk Mountains a great amount of Paleogene sediment was eroded at the beginning of the Miocene, whereas on the eastern and southern margins the denudation was less significant. In Early Miocene times rhyolite tuff and terrestrial beds were deposited locally, but it is difficult to estimate their thickness due to a subsequent Middle Miocene denudation event.

The following Middle Miocene dacitic pyroclastic rocks filled the eroded, karstic surface of the Paleo-Mesozoic formations of Bükk Mountains, but there is no data on the thickness of the post-Karpatian burial.

In the Late Pannonian (8–2.4 Ma), a sediment transport direction from north to south was proved in the southern margin of the Bükk Mountains (Bérczi et al. 1988).

Considerable erosion of the cover sediments and the development of the deep karst beneath the Fennsík (Bükk Plateau) Parautochthon started at the Pliocene/Pleistocene boundary (Hevesi 1980).

Thus the amounts of burial in the following three time intervals may be subject of speculation: pre-40 Ma (Late and post-Cretaceous uplift until E_3 sedimentation); 35–21 Ma (E_3–M_1 sedimentation with minor periods of variable denudation); and 16–2 Ma (M_2–Quaternary burial).

Additional important information can be concluded from the track length spectra. The major (longer) groups of tracks show a small, but not negligible, shortening. This refers to a very young, low-temperature overprint that was presumably related to very late termination of the burial.

Using the thermal modeling procedure of Willett (1992), several possible *tT* paths satisfying the geological constraints outlined here were obtained as the result of several hundred modeling runs (Fig. 9). All the acceptable *tT* paths need burial temperatures of about 50° C in the course of the 15–5 Ma period. As some small remains of Middle Miocene (Karpatian) tuffaceous sediments were preserved on top of the Bükk Plateau (Balogh 1964, see Fig. 7), the 'missing thickness' must have been post-Karpatian sediments. Five million years was chosen as the end of the warming up period, because the general trend observed in the evolution of the Pannonian Basin suggests that the erosion of the Late Miocene cover sequence of the Bükk Mountains could have started in Late Pannonian times (7–5 Ma; Horváth et al. 1988). The seismic sections across the southern marginal basin of the Bükk Mountains show nearly uniform southwards tilting of all Cenozoic isochron horizons (Tari 1988). This also refers to a Late Pannonian uplift of the Bükk's interior. Thus it can be concluded that the post-Karpatian burial of the Bükk Mountains was significant; according to estimations, at least 1 km of thick sedimentary pile was deposited on, and eroded after a short period from, the Bükk Plateau.

Conclusions

1. Resetting of the K-Ar system in the $<2\ \mu m$ grain-size fraction and the annealing of the fission tracks in zircon were approached only in the 'epizonal' and the high-temperature 'anchizonal' rocks. The effects of the detrital white micas are detected even in these rocks: the K-Ar ages of the metavolcanites devoid of

detrital muscovite are slightly, but systematically, younger than those of the clastic metasediments. In these rocks the illite-muscovite K-Ar and the zircon fission track ages are interpreted as cooling ages, i.e. as minimum age values of the regional metamorphism.
2. Zircon fission track ages of the medium and low-temperature 'anchizonal' and 'diagenetic' rocks do not date regional alteration; these data may refer to the source rocks of the detritus or can be considered as mixed ages without clear geological meaning. Of the illite-muscovite K-Ar data of these rocks, only the minimum values can be used for the rough estimation of the age of metamorphism.
3. The regional metamorphism of the Paleozoic and Mesozoic formations of the whole Bükkium can be dated between the 'eo-Hellenic' (Late Jurassic–Early Cretaceous, 160–120 Ma) and the Austrian (boundary of the Early and Late Cretaceous, 100–95 Ma) phases. The 'eo-Hellenic' phase is characterized by the subduction of the Dinarides and the Hellenides (Jacobshagen et al. 1976), whereas the Austrian phase is connected to the main folding and overthrusting (crustal thickening) processes in the Inner Carpathian realm. The concordant K-Ar and zircon fission track ages between 77 and 98 Ma in the eastern part of the Fennsík Parautochthon of the Bükk Mountains show that temperatures of >200° C also existed during the Subhercynian (90–85 Ma) phase.
4. In agreement with the earlier statements about the age relations of the regional metamorphism based solely on petrological, stratigraphic and tectonic data, there is no geochronological evidence for a pre-Alpine (Hercynian) metamorphic event.
5. The recently exhumed Paleozoic and Mesozoic formations reached the surface not much earlier than 40 Ma ago. The integrated thermal effect of the Paleogene and Neogene burial periods was significant. The thickness of the Middle Miocene-Pannonian burial reached or exceeded 1 km.

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