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NEOGENE CALC-ALKALINE INTRUSIVE MAGMATISM OF POST-COLLISIONAL ORIGIN ALONG THE OUTER CARPATHIANS: A COMPARATIVE STUDY OF THE PIENINY MOUNTAINS AND ADJACENT AREAS

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Abstract: A petrographical and geochemical analysis was carried out on intrusive rocks from the Pieniny and Moravian areas, with special attention to boron content and K-Ar radiometric ages.

The intrusions form medium- to high-potassium calc-alkaline andesitic suites, which are compositionally slightly different from each other and from the other calc-alkaline sequences in the Carpathian-Pannonian Region. No significant geochemical differences were observed within the different phase intrusions in the Pieniny areas. However, there is a slight difference in major and trace element composition between the Moravian and Pieniny intrusions. The andesitic rocks in the Pieniny and Moravian area are enriched in large ion lithophile elements and light rare earth elements and depleted in high field strength elements, indicating a metasomatized mantle source of the parent magmas. The low boron concentration of the andesitic rocks in the Pieniny area is in the range measured in back-arc, intraplate basalts of the Bakony-Balaton Highland volcanic field, whereas the higher boron content of the Moravian rocks overlaps with that of the Western Carpathian andesites. This may indicates the heterogeneity of the mantle lithosphere below the areas, or indicates different magma evolution histories.

On the basis of the systematic geochronological study, the intrusive rocks along the Outer Carpathians can be divided on three groups, which overlap with each other temporally. The oldest magmatism occurred from 14.8 Ma to 11.0 Ma in the Uhersky Brod area, Moravia, which was followed by the emplacement of andesitic dikes and sills in the Pieniny Mts., south Poland (13.5–10.8 Ma). In the Pieniny area, two intrusive phases were distinguished. Partly overlapping with this area, but generally younger than this magmatism, the emplacement of the youngest intrusions is referable to the Poiana Botizei-Ţibleş-Toroiaga-Rodna-Bârgâu intrusive area, Romania, where magmatic activity started at ~11.8 Ma and terminated at 8.0 Ma.

Key words: Outer Carpathians, Pieniny Klippen Belt, Neogene intrusive magmatism, K-Ar ages, boron concentrations.

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INTRODUCTION

The geological record of the Pieniny Mts., in the Western Carpathians arc in SE Poland, is dominated by a variety of magmatic episodes that reflect a complex tectonic history. The Pieniny Mts. are made up of a composite unit, consisting of structurally deformed Mesozoic to Neogene sedimentary sequences. Lines of isolated intrusions can be traced along this chain both westwards into the Western Carpathians (Moravia) and south-eastward into the Eastern Carpathians (Romania), forming a 700-km-long "outer magmatic arc" parallel to the Carpathian arc (Fig. 1).

The systematic study of the Pieniny Klippen Belt, involving a demonstration of its highly composite nature and its structural and chronological division into units, was begun by Birkenmajer 1970's (Birkenmajer, 1970, 1978, 1981, 1983, 1984; Grochocka-Piotrowska and Kibitlewski, 1974; Youssef, 1978; Birkenmajer and Nairn, 1979; Bir-

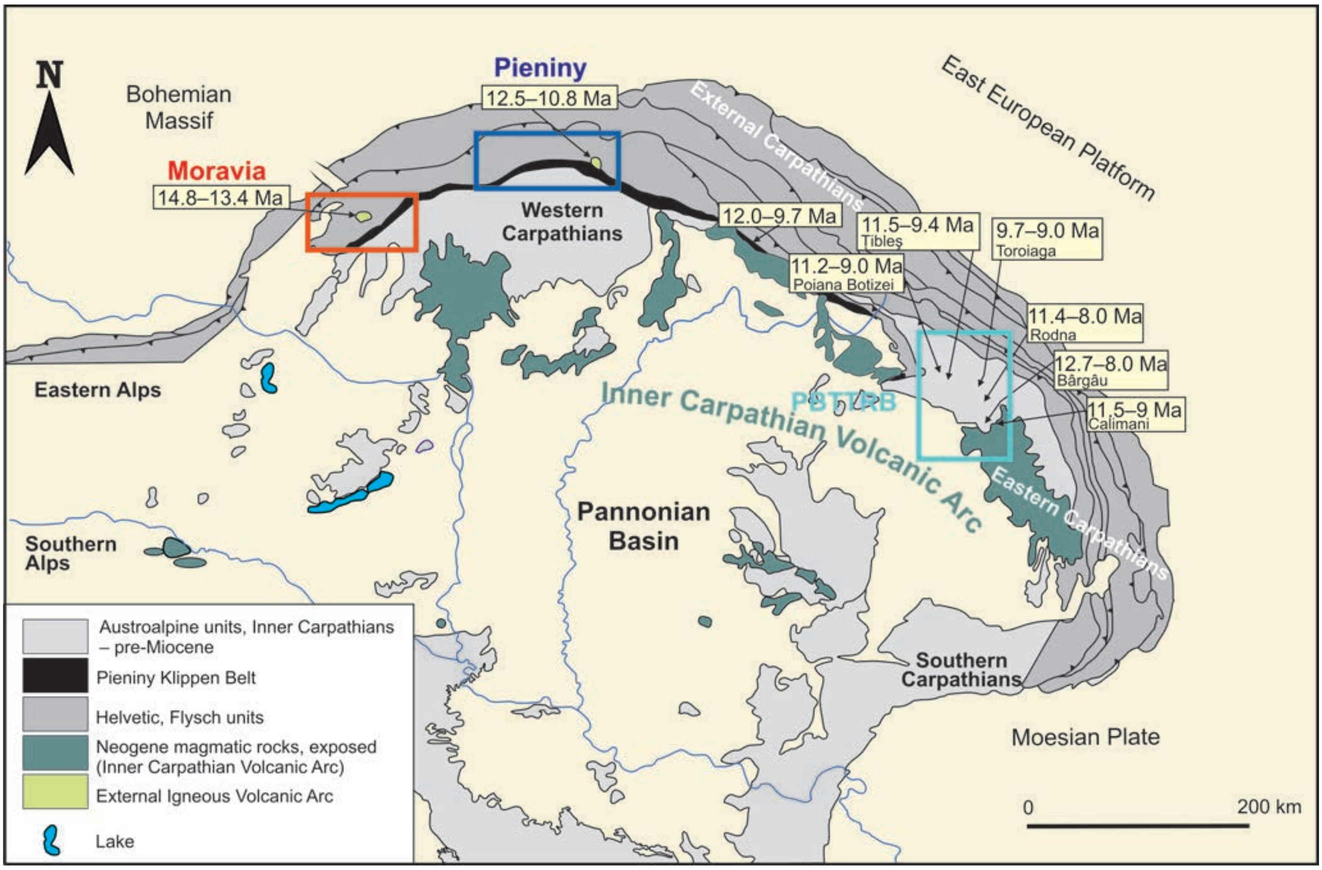

Fig. 1. Extent of Neogene–Quaternary calc-alkaline magmatic rocks and their age pattern along the Carpathian arc (modified after Pécskay and Gméling, 2010). PBTTRB – Poiana Botizei-Țibleş-Toroiaga-Rodna-Bârgâu intrusive area.

kenmajer *et al.*, 1979). On the basis of detailed mapping and field observations, Birkenmajer (1957, 1958) divided the intrusions into two major phases, which follow each other not only spatially, but also temporally (Birkenmajer, 1962).

Extensive petrographic work on the intrusions, exposed in the Pieniny Mts. (Birkenmajer, 1970, 1978, 1981, 1983, 1984), provided the basis for characterisation of the different volcanic associations in the adjacent areas. However, a better understanding of the relationship of intrusive magmatism to tectonism has been hampered by the lack of accurate and a systematic, radiometric data base.

Because of the discordant emplacement of these andesitic intrusions into the much older sedimentary sequences, radiometric dating is the only tool which makes possible the reconstruction of the evolution of this magmatic event.

The first K-Ar age determinations (Birkenmajer *et al.*, 1987) of these andesitic intrusions yielded widely-dispersed analytical ages (3.0–15.9 Ma), except for two meaningful geological ages (the isochron age; 12.6 Ma; Birkenmajer *et al.*, 1987 and the hornblende age; 13.5±1.0 Ma; Bukowski *et al.*, 1997). Detailed geochronological work began in 1998 in the framework of scientific cooperation between the Polish and Hungarian Academies of Sciences. K-Ar data, obtained on more than 40 representative samples, have played a crucial role in establishing the chronological framework for the intrusive magmatism and the tectonic evolution of the region (Birkenmajer and Pécskay, 1999, 2000). Since the publication of the geochronological papers, considerable advances have been made not only in documenting the distribution of Neogene calc-alkaline andesitic intrusions in space and time, but also in understanding the implications of this magmatic activity. Much of the progress has been achieved, owing to the collaborative efforts of geophysicists, structural geologists and geochemists, concerned with the relationship of intrusive magmatism to geodynamic processes (e.g., Seghedi *et al.*, 2004a, b, 2005; Pécskay *et al.*, 2006a, b; Trua *et al.*, 2006; Harangi and Lenkey, 2007; Harangi *et al.*, 2007; Nejbert *et al.*, 2012).

This paper is intended to provide a general summary of the Neogene calc-alkaline intrusive magmatic rocks of the Pieniny Mts., compared with similar intrusive rocks exposed in the "outer arc" of the Carpathian Pannonian Region (CPR). The authors discuss the significance of the new petrographical and geochemical data in relation to the origin of the Neogene calc-alkaline intrusive rocks, which are generally associated with much older sedimentary and/or metamorphic rocks.

GEOLOGICAL SETTING

The CPR had a very complex tectonomagmatic evolution during the Cenozoic as a consequence of the escape and rotation of Alcapa (Alp-Carpathian-Pannonian) and Tisza

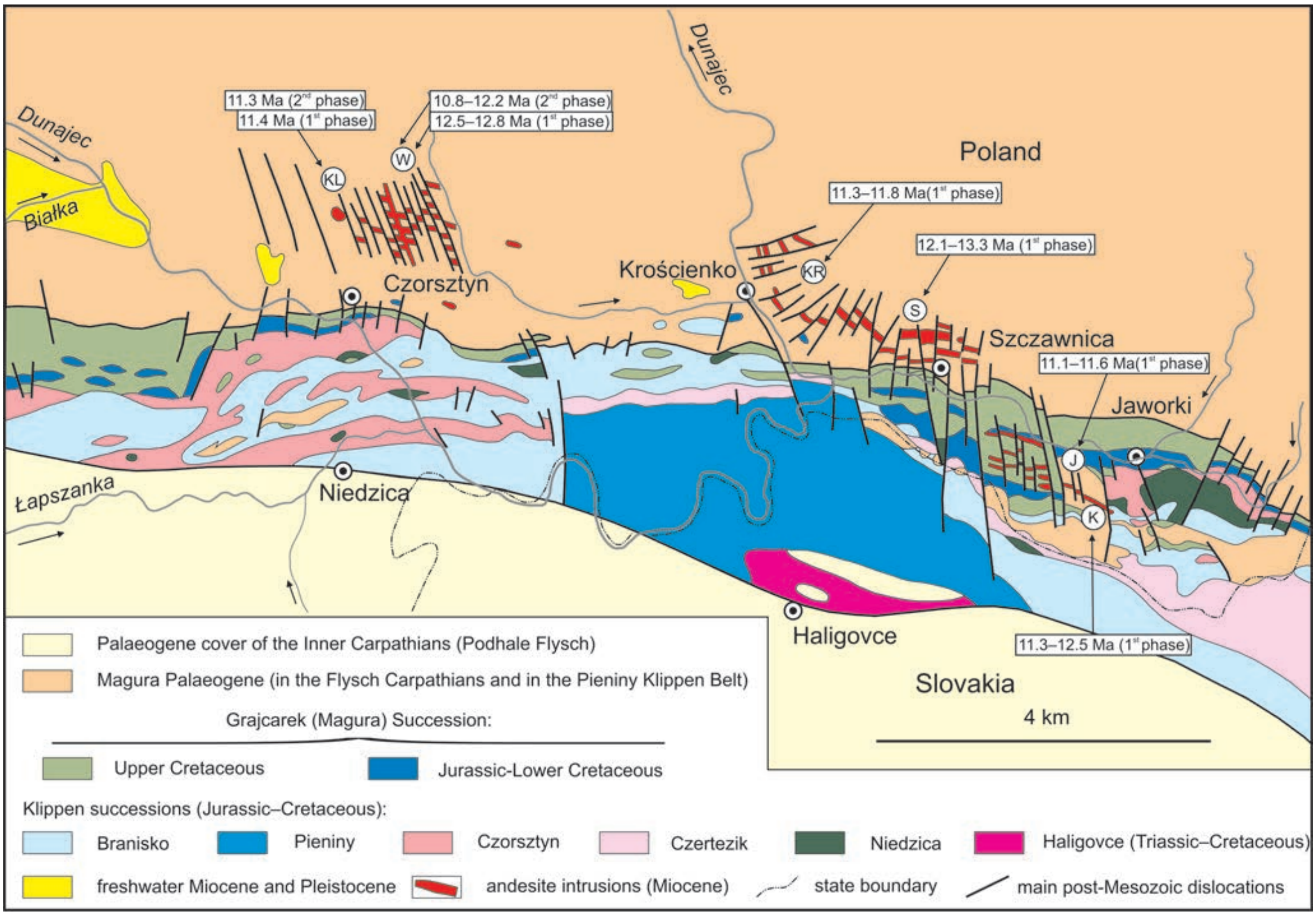

Fig. 2. Extent of the Pieniny Andesite Line intrusions in the Pieniny Mts. (modified after Birkenmajer and Pécskay, 1999). Abbreviations: KL – Kluszkowce; W – Mount Wżar; Kr – Krościenko; S – Szczawnica; J – Mount Jarmuta; K – Mount Krupianka.

microplates from the Alpine collisional zone. Their contemporaneous extension and collision with the European Plate acted at different times in different places. The large-scale (~400–600 km) extrusion of the microplates was triggered by the northward motion of the Adriatic microplate, the subduction and roll-back of the Magura Ocean (Kázmér and Kovács 1985; Csontos, 1995; Márton and Márton, 1996; Márton and Fodor, 2003; Csontos and Vörös, 2004). These plate-tectonic events were accompanied by arc-, back-arc and, intra-plate calc-alkaline and alkaline magmatism (Harangi *et al*., 2001, 2007; Seghedi *et al*., 2004a, b, 2005; Lexa *et al*., 2010; Seghedi and Downes, 2011). Volcanic and/or intrusive activity in the Inner Carpathian Volcanic Arc (Fig. 1) took place from 21 Ma until 0.1 Ma, with a distinct migration in time from west to east (Pécskay *et al*., 1995b; 2006a, b; Lexa *et al*., 2010).

The dominantly andesitic intrusive rocks of the “External Intrusive Volcanic Arc” (EIVA; Fig. 1) form an external arc, parallel to the northern and north eastern bend of the Western and North-Eastern Carpathians. The main magmatic fields (from west to east) are in Moravia and Pieniny in the Western Carpathians and in Poiana Botizei, Ţibleş, Toroiaga, Rodna, Bârgâu and Calimani in the Eastern Carpathians. These magmatic intrusions in the Western Carpathians follow the Pieniny Klippen belt or intrude into the external flysch zone of the Carpathians and are in a transitional position between the CPR and the Central European Volcanic Field. Therefore, their relationship to the subduction and extension in the CPR is controversial. Nejbert *et al*. (2012) proposed that the Moravian intrusions can be seen as transitional between the rift-related, alkaline magmatism of the foreland and the dominantly calc-alkaline magmatism of the Carpathian–Pannonian region. They suggested that the primary magmas of the Moravian and Pieniny intrusions were formed in a metasomatized mantle lithosphere. This metasomatism could have been related to an ancient subduction in the case of the Moravian intrusions and to deep mantle sources in the case of the Pieniny intrusions. Trua *et al*. (2006) suggested that the delamination process of the European plate that occurred during Middle Miocene in this sector of the Carpathian orogen should have favoured partial melting of metasomatized amphibole-bearing veins within the lithospheric mantle of the overriding Alcapa micro-plate.

Geology of the Pieniny intrusives

At the Slovakia-Polish border, products of intrusive activity form approximately a 20-km-long belt, called the Pieniny Andesite Line (PAL; Fig. 2). The intrusions have an *en échelon* relationship to the Dunajec Fault Zone (Birkenmajer, 1979), which is part of the NW-SE-oriented, mantle-

Table 1

Petrographic features of rock samples from Mount Wżar, and Mount Jarmuta (Pieniny Klippen Belt)

Sample name	Location	Rock type	Texture	Rock forming minerals	Hydrothermal/ contact metamorphic features	Comment
Intrusive rocks						
40P	Mt. Wżar/ Czorsztyn	1st phase amphibole-pyroxene andesite	micro-porphyritic	plagioclase (30%), pyroxene (15%), amphibole (10%), magnetite (5%), groundmass (volcanic glass, plagioclase 30–40%)	no	rich-in xenoliths, useful for K-Ar age dating
35P		2nd phase amphibole-pyroxene andesite	glomero-porphyritic	plagioclase (30%), amphibole (20%), pyroxene (5%), magnetite (3%), quartz (2%) groundmass (volcanic glass, plagioclase 30–40%), apatite (<1%)	no	useful for K-Ar age dating
30P		2nd phase amphibole-pyroxene andesite	porphyritic	plagioclase (30%), amphibole (20%), pyroxene (10%), magnetite (5%), groundmass (volcanic glass, plagioclase, goethite; 30–40%), apatite (<1%)	no	
Country rocks						
36P	Mt. Wżar/ Czorsztyn	allodapic limestone	wackestone	calcite, intraclasts: microcrystalline arenite, extraclasts: angular quartz crystals	no	not useful for K-Ar age dating
37P		mudstone	thin-bedded, laminated pelite	clay minerals, fine-grained muscovite, quartz	no	
39P		recrystallized, extraclast-rich limestone	equigranular crystalline	calcite (70%), extraclasts: quartz, muscovite; recrystallization: wollastonite.	contact effect of the andesite dykes is recognizable (recrystallized, wollastonite is present)	
39W		sandstone	non-bedded, well sorted, clastic	plagiocalse, muscovite, carbonate crystals, cemented by calcite	no	
41P		mudstone	well sorted, slightly foliated	clay minerals, muscovite, quartz, carbonate domains	no	
47P	Mt. Jarmuta	mudstone	non-bedded clastic, intraclast-rich	clay minerals, muscovite, quartz, carbonate domains	chalcedony veining may indicate contact metamorphism	
46P		mudstone	non-bedded clastic, intra-, and extraclast-rich	clay minerals, muscovite, quartz, carbonate domains, extraclasts: limestone	abundant fine muscovite crystals may indicate weak recrystallization as a result of contact metamorphism	
45P		aleurolite-sandstone	foliated	quartz, clay domains, cemented by calcite	no	

rooted structure, called the Kraków-Myszków Fault Zone. The dextral strike-slip movement along this zone provided a migration path for the magmas to intrude from the mantle into the upper crust (Nejbert *et al*., 2012). The intrusions were emplaced into the Lower Jurassic–Upper Cretaceous marine sedimentary rocks of the Grajcareck Unit and its Palaeogene sedimentary cover and the Upper Cretaceous–Eocene flysch rocks of the Magura Nappe (Fig. 2).

Emplacement of the intrusions took place in two phases (Birkenmajer and Pécskay, 1999, 2000)· First-phase intrusions form dyke swarms subparallel to the Pieniny Klippen Belt and were faulted during Sarmatian times. The 2nd phase intrusions are pronounced in the westernmost part, but also are present in the eastern segment of the PAL. The 2nd phase intrusions follow transversal faults that cut the 1st phase andesites. On the basis of the field observations and the radiometric data, the emplacement of these andesitic intrusions unambiguously post-date the early Miocene compression, responsible for the intense folding of the Klippen Belt and flysch zones (Birkenmajer and Pécskay, 1999, 2000).

Previous studies (Trua *et al*., 2006; Gméling *et al*., 2008; Nejbert *et al.,* 2012) discussed the petrographical and geochemical characters (major- and trace-element and isotopic compositions) and provided various models for the Pieniny intrusions. The intrusions form medium to high-K calc-alkaline rock suites and are different from each other and from other calc-alkaline sequences of the CPR. Geochemically, they are transitional between rift-related alkaline magmatism and the calc-alkaline magmatism of the CPR. The contrast between the dominance of basaltic andesites on the western side of the “outer arc” and more acid (dacitic and rhyolite) on south-eastern side is consistent with the changes in the properties of the lithosphere. The variable degree of iron enrichment shown by these intrusions may be due to differences in fractionation conditions, but variations in potassium and some of the trace elements may mean a heterogeneous magma source (Seghedi *et al*., 2004a). The Pieniny rocks are enriched in highly incompatible trace elements (LILE and LREE) and depleted in high-field-strength elements (HFSE), compared to the primitive mantle, similar to most Carpathian Arc magmas (Trua *et al*., 2006).

METHODS

Determination of boron content was carried out, using the prompt gamma activation analysis (PGAA) facility at the Budapest Research Reactor (Hungary). PGAA is especially useful for analysing whole-rock boron concentrations. In contrast to other geoanalytical methods, with PGAA sample preparation procedures are not needed and hence contamination problems are eliminated (Anderson and Kasztovszky, 2004). Accuracy of boron analyses by PGAA has been checked by measurements of geological reference materials (Gméling *et al*., 2005; Gméling *et al*., 2014). The B data have a small relative uncertainty (1–1.5%).

RESULTS AND DISCUSSION

New petrographic data of the intrusives and their country rocks in the Pieniny Mountains

Samples of the 1st and 2nd intrusive phases and their country rocks were collected in two quarries at Mt. Wżar near Czorsztyn and at Mt. Jarmuta at Malinów (Fig. 2). Petrographic features (modal composition, rock texture and alterations) are summarized in Table 1.

The intrusive rocks were previously classified as two main groups: the amphibole andesites and amphibole-augite andesites (Małkowski, 1921, 1958). Both 1st and 2nd phase andesite samples – studied recently – are amphibole-augite andesites, therefore the classification for the study area suggested above does not agree with the temporal (1st and 2nd phase) distinction of the intrusions.

The 1st phase intrusive rock is an amphibole-pyroxene andesite showing microporphyritic texture (Fig. 3A). The most abundant phenocrysts are plagioclase, amphibole and clinopyroxene. Plagioclase is resorbed by the groundmass that crystallized to smaller plagioclase crystals. On the basis of optical observations, plagioclase compositions range from labradorite to andesine. The amphibole has been identified optically as hornblende (Fig. 3A). On the basis of optical observations, the clinopyroxene phenocryst is augite. Amphibole and plagioclase are zoned: the core of the crystals was formed in an early phase of the crystallization of the melt, whereas the outer zones represent a late crystallization event. Minerals in the groundmass are magnetite and submicroscopic crystallites. The groundmass is inhomogeneous (dominantly pilotaxitic, locally hyalopilitic). The zoned phenocrysts and the inhomogeneous texture of the groundmass indicate that the rock experienced a multiphase crystallization history and that the crystallization rate during intrusion and cooling was not uniform. Three different xenoliths with diameters of up to 1.5 mm were observed in the 1st phase andesite: 1) dark, rounded, very fine-grained, recrystallized inclusions that originated probably from the metasedimentary footwall; 2) oriented plagioclase-rich fragments in a glassy groundmass, which originated from the margin of the andesite dykes that experienced rapid cooling; and 3) a granitoid inclusion with partially molten plagioclase and altered biotite. Alteration of the biotite is the result of the reaction of the biotite and plagioclase. This latter inclusion probably comes from the crystalline basement.

Second-phase intrusions have glomeroporphyrytic or porphyritic texture. Phenocrysts, are zoned plagioclase (andesine-labradorite; Fig. 3B), zoned hornblende and clinopyroxene (augite). The various hialopilitic-pilotaxitic textures of the groundmass (Fig. 3C) prove the multi-phase crystallization process of the dykes. The porphyritic texture of sample 30P indicates that the phenocrysts were already crystallized by the time of the intrusion and the fine-grained groundmass crystallized relatively rapidly during the intrusion of the magma.

Several secondary hydrothermal processes in the 1st phase dykes and their country rocks, associated with the 2nd phase magma intrusion have been distinguished by Youssef (1978) and Trua *et al*. (2006).

Hydrothermal alteration and ore-mineralization in the eastern part of the PAL (Krościenko, Szczawnica, and Mt. Jarmuta) of the 1st phase intrusions was reported by Birkenmajer *et al*. (2004). The veins in these regions experienced three phases of hydrothermal alteration. During the 1st high-temperature hydrothermal phase, biotite with high Cl contents, intergrown with quartz and Cl-apatite were formed. The high Cl content of the biotite is evidence for a secondary origin and thus suitable for K-Ar dating of the hydrothermal process (Birkenmajer *et al*., 2004). During the 2nd low-temperature, hydrothermal phase, the andesite was propilitized (albite-epidote-chlorite alteration). Ore minerals (e.g., pyrrhotite, pyrite, chalcopyrite and electrum) formed in this phase. During the 3rd hydrothermal phase, carbonatization of the andesite was caused by cool carbon-dioxide-rich waters (Birkenmajer, 1958; Małkowski, 1958). During the 4th hydrothermal phase, low-pH and low-temperature fluids replaced the primary rock-forming minerals (e.g., pyroxene) by Fe-oxides and the 2nd phase sulphides; by covellite and marcasite.

Except for the opacite rim of amphibole – indicating deuteric alteration – no secondary hydrothermal or thermal alteration was observed in the 1st phase samples. In some 2nd phase intrusions, weak goethitic alteration, a result of oxidation of the primary silicate phases, is present.

The country rocks studied are various clastic (mudstone-sandstone) and carbonate rocks (limestone). Contact metamorphic effects of the intrusive rocks was observed only in three samples, in form of chalcedony veining (47P; Fig. 3D), the occurrence of wollastonite (39P; Fig. 3E) in limestone and the occurrence of muscovite as a result of recrystallization in mudstone (46P; Fig. 3F).

Contact-metamorphic alteration of the sedimentary country rocks was observed by Birkenmajer *et al*., (2004). In the contact-close zones, wollastonite, diopside, pigeonite, and cristobalite, in the distal zones wollastonite, diopside and garnet occur. Hydrothermal circulation around the dykes resulted in the formation of argillitization, formation of carbonate and ore minerals (pyrite, pyrrhotite, chalcopyrite).

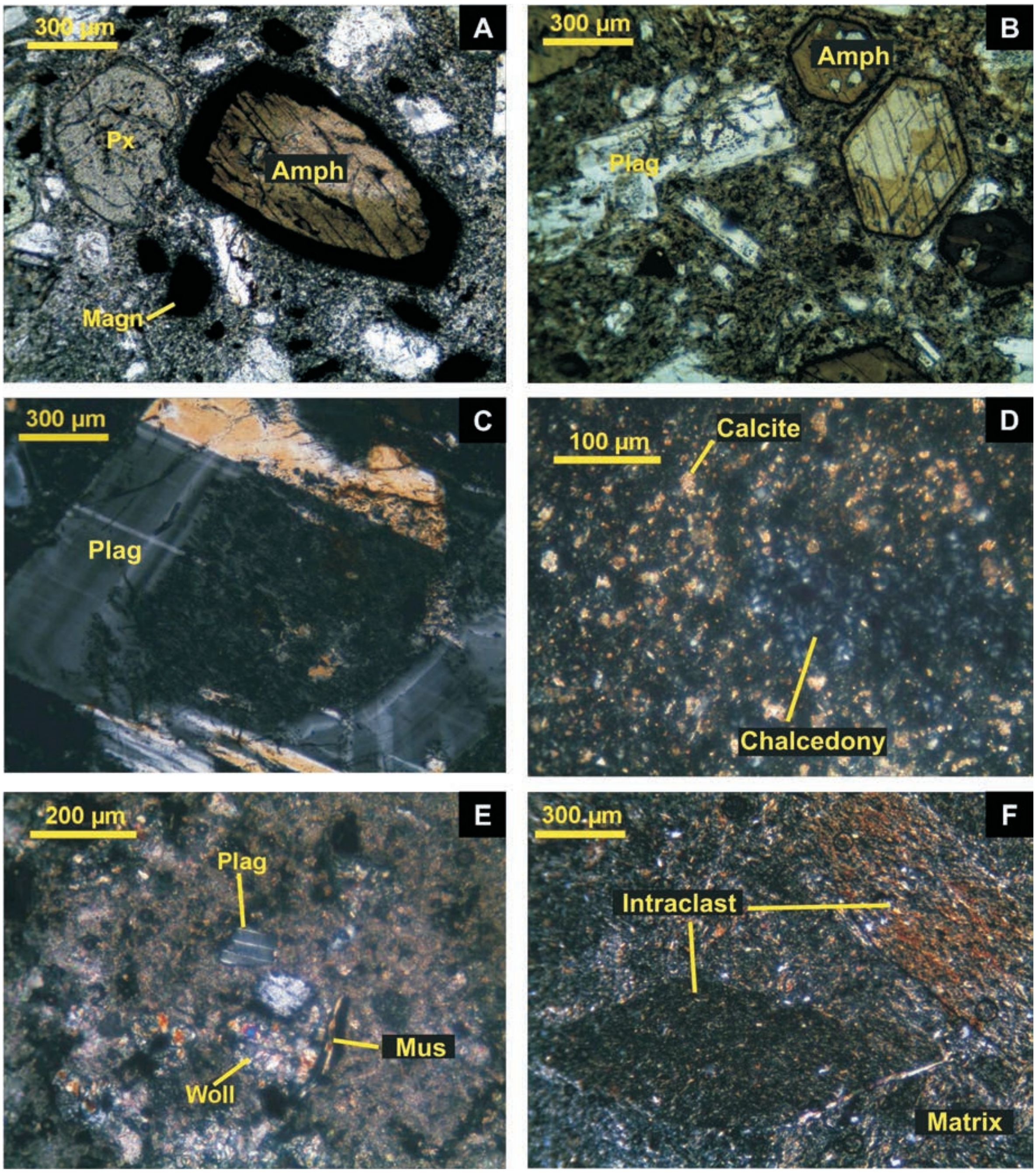

Fig. 3. Photomicrographs of andesite dikes **A.** 1st phase andesite dyke: zoned, euhedral amphibole with opacitic rim, subhedral pyroxene and plagioclase phenocrysts in a hyalopilitic-microporphyric groundmass. **B.** Euhedral amphibole and plagioclase in a hyalopilitic-pilotaxitic groundmass. **C.** Large, euhedral plagioclase with glass-rich core. The outer zones show oscillatory zoning using crossed polars, as evidence of variation in the composition. **D.** Irregular chalcedony domain in limestone (Sample 47P). **E.** Wollastonite-rich domain in limestone (sample 39P). **F.** Muscovite-rich mudstone with intraclasts.

Geochemistry of intrusive rocks in the Pieniny Mountains and in Moravia

The geochemical data are summarized in the Appendix.

With one exception, the rocks studied can be classified as medium-K, calc-alkaline, ranging from basaltic andesites to andesites. The 1st phase intrusions show greater variation in SiO_2 content (Fig. 4A, B). No variation in trace element composition of 1st and 2nd phase intrusions can be observed in Figure 4C.

In general, the Pieniny rocks are enriched in highly incompatible trace elements (LILE, LREE), and depleted in high-field-strength elements compared to the primitive mantle, similar to most Carpathian arc lavas (Fig. 4D). The LILE enrichment indicates that the melting occurred in the metasomatised subcontinental lithospheric mantle. The metasomatism can either be related to the Neogene subduction of the Magura Ocean or to an older subduction event.

The positive Pb anomaly reflects a component, derived from continental crust in the parent magma.

The relative freshness of most of these samples is supported by their low LOI (less then 3 wt.%; Appendix 1).

The B content of the Pieniny andesites is between 2.97 and 29.5 µg/g. This range overlaps with the B content of the andesitic volcanic rocks in the Western Carpathians (Fig. 5A). The lower values of the Pieniny area are more in the range measured in back-arc, intraplate basalts of the Ba- kony-Balaton Highland volcanic field (1.6–12.9 µg/g; Gméling *et al.*, 2007). Using the LILE/HFSE and LILE/ REE ratios the fluid transferred from the subducted slab and sediments into the overlain mantle wedge can be monitored (Vroon *et al.*, 1993). On the plot of B/Sm *versus* Ba/Sm (Fig. 5B) two trends can be seen: the high Ba/Sm ratios indicate addition of fluids from the subducted slab, and the high B/Sm ratios indicate involvement of fluids in the magma genesis from sediments. High La/Nb ratios and variable B/Nb ratios (Fig. 5C) are indicative for metasomatic processes of the lithospheric mantle during subduction (Ishikava and Tera, 1997).

The average B content of the Moravian rocks examined are slightly higher (~15 and 7 µg/g respectively). However, some Pieniny rocks show extremely high values of around 20 µg/g (Fig. 5A). The relatively higher B contents of the Moravian rocks correlate with the higher K_2O content of these rocks compared to the andesites in the Pieniny Klippen Belt. The B data of the Moravian rocks overlap with the B content of Western Carpathian andesites (11.1–29.8 µg/g). The higher K_2O/Sm ratio of the Moravian rocks, refers the metasomatic origin of the fluids originating from the crust. The fluid added to the source of the Pieniny rocks more probably originated from the subducted sediments, causing elevated B/Sm ratios (Fig. 4B). The flysch sediments from the region examined also show high B/Sm ratios (Gméling, 2010).

There are two 1st phase samples from the peak of Mt. Wzar with anomalously high boron content (29.5 and 18.9 ěg/g respectively) and B/Gd ratios (4–8) (Fig. 4A, E). As these 1st phase intrusions are in spatial contact with 2nd phase intrusions, probably the hydrothermal fluid flow associated with the 2nd phase intrusions geochemically modified the 1st phase intrusions. Hydrothermal activity enriched the intrusions in fluid-mobile elements, especially the boron and the lead content of the rocks.

Effect of the fluids on the K-Ar ages based on the boron geochemistry of some Pieniny intrusions

Thermal/hydrothermal processes may cause secondary alteration of the rock, forming minerals that can affect the K-Ar radiometric age of the rock through addition or loss of K and Ar. Examination of the B content as a function of the K-Ar age can be indicative of the role of fluids on the radiometric age of the rock. No systematic relationship between B/Gd ratios vs. H_2O content of the 2nd phase intrusions can be detected in Fig. 5D. Some 1st phase intrusions, however, show a slight increase in B/Gd ratios. In the western part of the Pieniny Mts. (Mt. Jarmuta, Krościenko, Krupianka) only weak evidence of fluid addition can be detected, whereas in the western part (Kluszkowce, Mt. Wżar) andesites show a slightly increasing fluid involvement with time that may have affected the K-Ar age of the rocks (Fig. 5E).

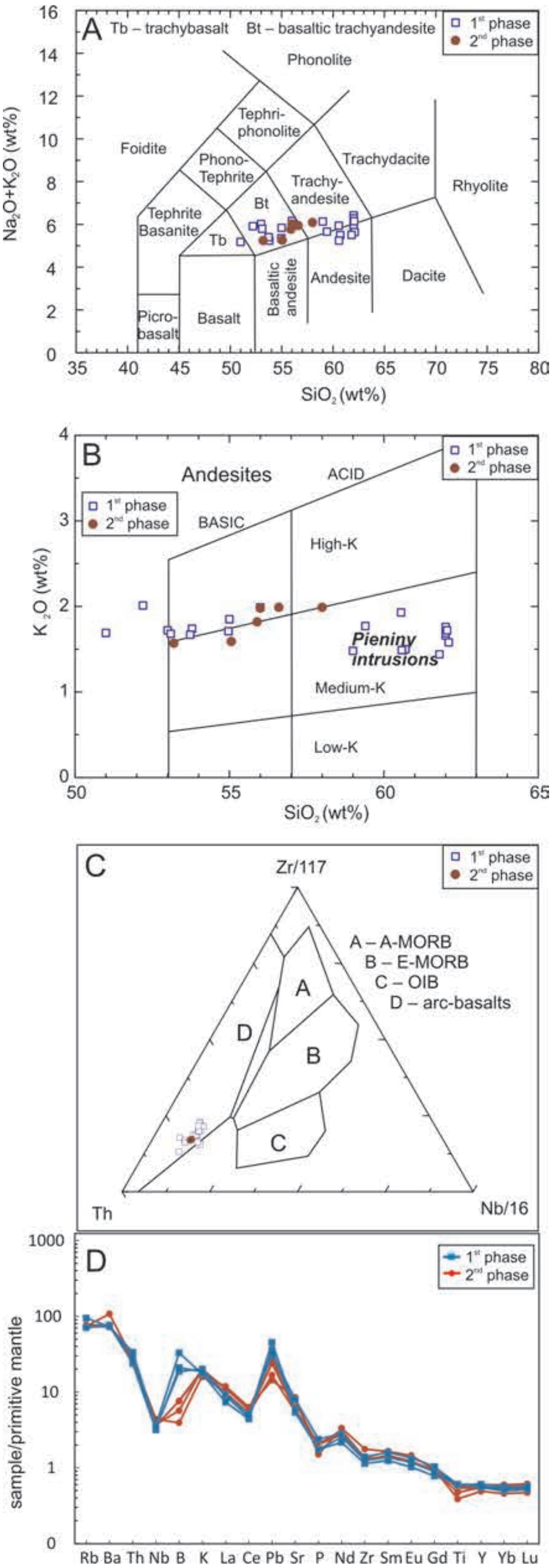

Fig. 4. New geochemical data of intrusions from the Pieniny Mts. **A.** Total Alkali–Silica diagram (TAS: Le Maitre, 1989). **B.** K_2O (wt%) vs. SiO_2 (wt%) diagram (Peccerillo and Taylor, 1976) of intrusive rocks from the Pieniny Mts. **C.** Th-Zr/117-Nb/16 (ppm) trace-element distribution diagram. **D.** Primitive-mantle-normalized element variation diagrams for Pieniny Mts. samples. Normalizing values and E-MORB data from Sun and McDonough (1989).

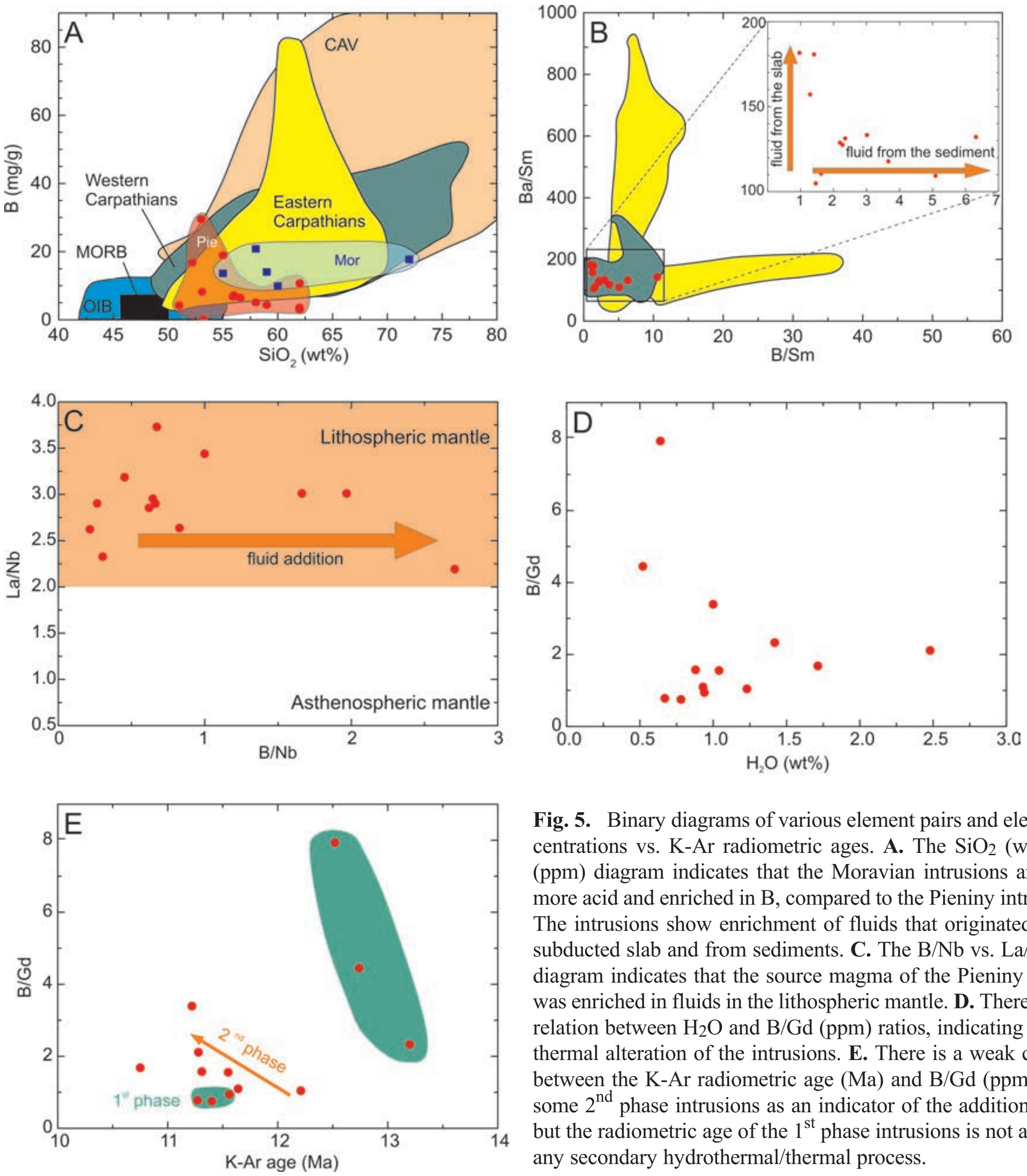

Fig. 5. Binary diagrams of various element pairs and element concentrations vs. K-Ar radiometric ages. **A.** The SiO_2 (wt%) vs. B (ppm) diagram indicates that the Moravian intrusions are slightly more acid and enriched in B, compared to the Pieniny intrusions. **B.** The intrusions show enrichment of fluids that originated from the subducted slab and from sediments. **C.** The B/Nb vs. La/Nb (ppm) diagram indicates that the source magma of the Pieniny intrusions was enriched in fluids in the lithospheric mantle. **D.** There is no correlation between H_2O and B/Gd (ppm) ratios, indicating no hydrothermal alteration of the intrusions. **E.** There is a weak correlation between the K-Ar radiometric age (Ma) and B/Gd (ppm) ratios of some 2nd phase intrusions as an indicator of the addition of fluids, but the radiometric age of the 1st phase intrusions is not affected by any secondary hydrothermal/thermal process.

K-Ar geochronology of emplacement of the intrusive rocks along the "EIVA"

The intrusive magmatic formations are important geological units within the Neogene–Quaternary calc-alkaline volcanic activity of CPR, which generally offer views on the roots of the magmatic structure. Furthermore, the distribution of the volcanic rocks shows a strong correlation with local fault structures, implying a strong genetic relationship between magmatic activity and Neogene tectonism. It is widely accepted that there is a genetic connection between the volcanic and intrusive rocks (Seghedi *et al*., 2004a, 2005). This conclusion has been also confirmed by accurate radiometric dating of the intrusions and the corresponding lavas (e.g., Pécskay *et al*., 2006a).

The extended range of the K-Ar dates (15.0–8.0 Ma) presented in Figure 6 support recognition of spatial groups along the "EIVA". There are some temporal trends in the distribution of rock types along the CPR. The K-Ar ages indicate that the intrusive magmatism started at about 15.0 Ma ago (Badenian) and continued through the Sarmatian, terminating at about 8.0 Ma (early Pannonian; Pécskay *et al*., 1995b, 2006a; Fig. 6A).

The oldest intrusive rocks, forming dikes, sills and laccoliths, are exposed on the NW side of the "EIVA", Western Carpathians, Moravia (Fig. 6A). These intrusions yielded dates that range from 14.8 Ma to 11.0 Ma (Pécskay *et al*., 1995b, 2006a). On the basis of K-Ar data, three pulses of intrusions can be assumed in this region; 14.8–14.4 Ma; 13.5–13.4 Ma and 12.7–11.0 Ma, respectively. The small outcrops of andesite dykes in western Slovakia (Fig. 1) are the eroded remnants of dike swarms. One representative sample taken from these dikes at the Horné Srnie locality gave 11.8 Ma (Fig. 1), which is compatible with the youngest intrusions cropping out in Moravia (Pécskay *et al.*, 1995b).

In the Pieniny Mts., outcrops of intrusions are spatially restricted to the maximum bend and northward extension of the Pieniny Klippen Belt (Birkenmajer, 1984; Fig. 2). Their size and shape are related to intrusive rock type and relative time of emplacement: the earlier/older (1st phase), more basic intrusions are much larger than the later varieties (2nd phase).

The most preferable age interval for the Neogene intrusive magmatism in the Pieniny Mts. is from 13.3 Ma to 10.8 Ma (Sarmatian). The oldest andesitic intrusions are exposed near Szczawnica and yielded ages that range from 13.3 Ma to 12.1 Ma (Figs 2, 6A). The 1st phase intrusions, mainly dikes, subordinately sills, are the most frequent magmatic forms. Their distribution shows a strong correlation with local fault structures (see Fig. 2; e.g., Birkenmajer, 1979, 1984). K-Ar dates on representative samples belonging to the 1st phase show a range of 13.3 Ma–12.1 Ma. The apparent ages younger than 12.0 Ma are related to some secondary effect, which caused Ar loss (e.g., these rocks were rejuvenated by this secondary activity; Birkenmajer and Pécskay, 1999, 2000).

The chronological order of emplacement of the various intrusions deduced from field evidences is not always reflected in detail by the age data. It seems that an incremental heat effect occurred, owing to the closely spaced pulses of magma, superimposed on one another. The most reliable K-Ar ages, especially from the western side of the Pieniny Mts. – correspond to the climax of this intrusive activity, which was marked by the emplacement of the largest intrusions, belonging to the 1st phase.

On the other hand, it is also striking that on the basis of the radiometric ages, some amphibole and plagioclase mineral fractions separated from the andesitic intrusions gave much older ages than the whole-rock age of the same representative sample. One might consider that the older ages are the consequence of some excess Ar, caused by the presence of a xenolith or xenocrysts in the analysed rock samples. It is well known that amphibole phenocrysts crystallise quite early in relation to plagioclase during the cooling of magma; therefore, the crystallisation age of the amphibole and the age of emplacement of the intrusion (Birkenmajer and Pécskay, 2000) can be different. Nevertheless, for the sake of correct interpretation of these K-Ar ages, it would be necessary to identify all the xenoliths/xenocrysts, if they are true accidental xenoliths, derived from the early intrusive phases, or torn-off xenoliths from the country rocks.

According to the field observations of Birkenmajer (1984), the 2nd phase intrusions are spatially restricted to the westernmost part of the Pieniny Mts. However, the geochronological data do not confirm this geological interpretation (Fig. 2; Birkenmajer and Pécskay, 1999, 2000). The termination of the intrusive magmatism that occurred in the Pieniny Mts. has been precisely determined (10.75±0.46 Ma).

Neogene andesitic intrusive rocks are found also in spatial groupings in the Kapušany-Vinné complex of Eastern Slovakia. Their distribution also shows a strong correlation with local fault structures. K-Ar dates on these rocks are 12.2–11.7 Ma (Pécskay *et al.*, 2006). These radiometric ages, together with petrographical and geochemical features, link these intrusions with the similar rocks belonging to the 1st phase of the Pieniny Mts.

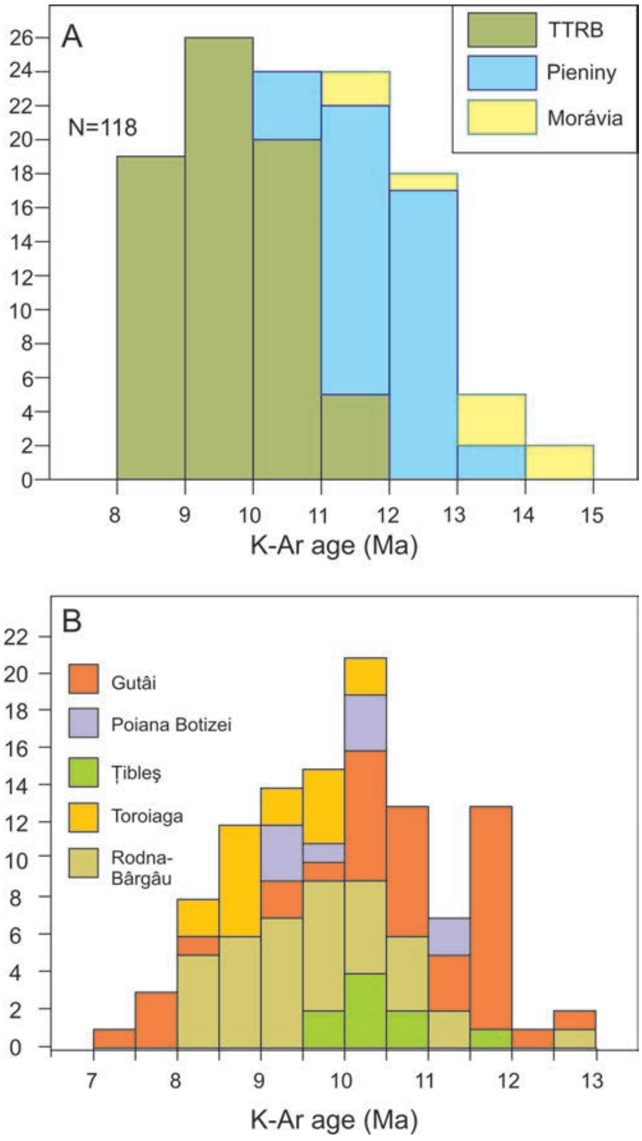

Fig. 6. Distribution diagrams of K-Ar radiometric ages **A.** K-Ar age distribution of the Extrenal Intrusive Volcanic Arc intrusions (PBTTRB, Pieniny, Moravia). **B.** K-Ar age distribution of the PBTTRB intrusions.

Close to the Kapušany-Vinné complex, areas of calc-alkaline andesitic rocks showing volcanic landform, generally forming composite volcanoes in the Vihorlát-Popriečny Mts., are similar in age (Kaličiak *et al.*, 1995).

A relatively small sill intruded into the flysch deposits, outcropping immediately to the northeast of the Siniak stratovolcano, Transcarpathia, yielded 11.8 Ma (Pécskay *et al.*, 2000). The chemistry and age of this rock are compatible with interpretation of the authors that this sample represents the same magmatic association belonging to the EIVA (Seghedi *et al.*, 2001).

The south-eastern groups of intrusions exposed between the Oas-Gutai Mts. and Calimani Mts., Eastern Carpathians (Poiana Botizei-Ţibleş-Toroiaga-Rodna-Bârgâu) are referred to collectively as PBTTRB (Figs 2, 6B; Pécskay *et al.*, 2006b, 2009). According to the detailed geochronology, they are the youngest manifestation of the EIVA to be considered as Sarmatian and Lower Pannonian in age (14.0–8.0 Ma; Pécskay *et al.*, 1995a, 2009) and form the third age group.

CONCLUSIONS

The K-Ar dates obtained on the intrusions along the EIVA indicate that predominantly medium to high-K calc-alkaline andesitic intrusive magmatism commenced about 15 Ma ago on the western side of the EIVA. During the period 15.0–8.0 Ma, this magmatism extended along the arc and terminated about 8 Ma ago on the south-eastern side of the EIVA, Romania. On the basis of the radiometric data and in accordance with the geological observations, it has been proved that this magmatism was episodic. Neither petrographical nor geochemical differences were found between the different phases of andesitic intrusion in the Pieniny Mts. However, K-Ar ages proved that magmatic activity in the Pieniny Mts. was a two-phase process.

The Neogene calc-alkaline intrusive magmatism commenced as a result of tectonic compression at a convergent plate boundary and was probably associated with a preceding period of subduction. Boron concentration analysis confirmed previous models indicating that the source region beneath the volcanic arc was heterogeneous. On the basis of various boron/incompatible element ratios, partial melting occurred in a previously metasomatized mantle wedge. Boron concentration analysis also proved to be a useful tool for interpretation of anomalously old K-Ar ages and detection of thermal/hydrothermal effects in the intrusive rocks.

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Appendix

Major- and trace-element data for Pieniny Mountains and Moravian samples

	Mt. Wżar 1st phase intrusions								Mt. Wżar 2nd phase intrusions					Moravian intrusions				
	98/13P	98/7P	98/6P	98/17P	98/12P	99/24P	99/21P	99/20P	98/8P	98/1P	98/5P	99/27P	99/25P	KPM-4/I	KPM-1	KPM-6	KPM-4/IIB	KPM-4/IIA
SiO_2	62.31	54.06	54.74	60.34	60.57	52.3	61.17	52.77	56.64	55.89	56.3	52.91	55.76	60	55	59	58	72
TiO_2	0.51	0.79	0.72	0.51	0.49	0.74	0.51	0.78	0.69	0.67	0.71	0.77	0.68	0.69	0.98	0.71	0.61	0.69
Al_2O_3	18.66	18.91	18.92	18.04	18.44	18.69	19.18	18.87	18.72	18.64	18.89	17.63	18.36	17.2	19.8	19.8	18	18.2
Fe_2O_3(T)	4.76	7.75	7.35	5.02	4.67	8.05	4.96	8.18	6.77	6.75	6.83	6.92	6.72	4.3	7.6	5.9	4.7	5.6
MnO	0.12	0.16	0.14	0.13	0.13	0.16	0.14	0.19	0.15	0.15	0.15	0.13	0.15	0.062	0.301	0.199	0.188	0.134
MgO	0.99	3.48	2.9	1.44	1.12	3.26	1.17	3.57	2.68	2.88	2.9	4.07	2.88	2.5	1.6		1.5	2.3
CaO	5.99	8.79	8.07	5.95	6.27	8.89	5.71	9.18	7.61	8.32	8.15	8.66	7.38	4.7	6.2	5.4	5.3	4.9
Na_2O	4.09	3.81	3.6	3.76	4.46	3.48	4.7	3.52	3.65	3.83	3.67	3.64	3.68	4.76	4.9	5	4.49	5.4
K_2O	1.54	1.7	1.89	1.56	1.88	1.67	1.66	1.85	1.8	1.58	1.71	1.54	1.64	3.19	3.01	3.17	3.72	3.95
P_2O_5	0.24	0.28	0.28	0.26	0.24	0.37	0.24	0.38	0.33	0.32	0.33	0.26	0.32					
LOI	1.26	0.62	1.24	3.45	2.19	2.7	1.06	1.04	1.78	1.41	1.24	4.31	3.17					
Mg#	0.31	0.48	0.45	0.38	0.34	0.45	0.33	0.47	0.45	0.47	0.46	0.55	0.47					
B	3.53	29.5	18.9	10.69	2.97	4.24	4.37	16.8	5.11	6.82	6.91	8.19	7.23	9.86	13.62	14.02	20.8	17.7
Cl	73	160	75	70	54	77	137	178	130	112	112	65	110	370	95	111	231	208
Nd		28	26	31	33	23	26			29	26	20	28	36		47	45	43
Sm	2.74	2.8	2.99	2.92	3.08	2.91	3.11	3.3	3.15	2.9	3.04	2.72	3.3	3.16	4.6	4.2	3.51	3.56
Gd	3.1	3.5	3.6	3.3	3.4	3.7	3.5	3.9	3.5	3.5	3.7	3.3	3.5	3	4.8	4.3	3.3	3.41
Cr	4	25	15	10	5				22	22	25	41						
Co	4	14	14	8	4	17	5	17	13	13	12	17	13					
Rb	54	52	53	52	58	47	61	69	56	52	51	42	52					
Sr	976	620	672	937	910	792	879	927	678	821	638	597	596					
Y	18	20.2	21	20.4	17.3	20.1	17.8	22.3	22	19.8	19.8	17.7	19.6					
Zr	169	109	120	165	170	108	173	134	129	124	121	110	121					
Nb	13.3	10.9	9.6	10.7	13.8	6.3	14.4	10.1	11.3	11	10.7	9.9	10.9					
Cs	1	3.4	2.7	6.1	1.3	1.3	2	5.2	1.8	1.5	1.3	1.4	1.1					
Ba	880	607	621	674	892	515	891	597	621	633	635	581	630					
La	38.6	23.9	28.9	36.8	36.2	23.5	33.5	30.4	36	31.4	31.6	26.1	31.6					
Ce	61.6	43	45.6	59.8	61.9	40.8	58	50.2	56.9	51.7	51.4	42.4	50.4					
Pr	8.41	5.17	6.27	8.39	7.35	5.73	6.84	6.89	8	6.95	6.92	5.8	6.88					
Nd	31.6	20.5	24.1	31.4	27.8	23.1	26.3	26.9	29.6	26	26.2	22.3	25.8					
Sm	5.6	4.25	4.7	5.72	4.91	4.91	4.93	5.46	5.62	4.82	4.98	4.36	4.89					
Eu	1.93	1.36	1.56	1.86	1.69	1.58	1.64	1.78	1.82	1.6	1.62	1.45	1.59					
Gd	4.52	3.72	4.25	4.59	3.95	4.49	3.99	4.95	4.9	4.39	4.39	3.88	4.3					
Tb	0.7	0.64	0.71	0.73	0.65	0.75	0.65	0.82	0.79	0.71	0.73	0.65	0.72					
Dy	3.61	3.66	4.02	3.95	3.37	4.18	3.47	4.39	4.41	3.93	4.04	3.58	3.94					
Ho	0.65	0.73	0.76	0.74	0.62	0.79	0.64	0.82	0.84	0.76	0.77	0.68	0.75					
Er	1.89	2.11	2.22	2.15	1.78	2.24	1.82	2.39	2.44	2.19	2.2	1.93	2.16					
Tm	0.269	0.305	0.329	0.315	0.264	0.328	0.272	0.346	0.357	0.327	0.326	0.278	0.325					
Yb	1.81	1.96	2.11	2.1	1.78	2.05	1.87	2.25	2.36	2.1	2.15	1.86	2.09					
Lu	0.277	0.304	0.314	0.321	0.264	0.308	0.27	0.34	0.361	0.324	0.322	0.272	0.32					
Hf	4.8	3.25	3.6	4.8	4.6	3.3	4.7	3.8	3.8	3.7	3.6	3.4	3.6					
Ta	0.96	0.68	0.74	0.82	0.92	0.53	0.96	0.78	0.9	0.86	0.85	0.76	0.85					
Tl	0.18	0.18	0.27	0.27	0.21	0.19	0.27	0.44	0.27	0.27	0.21	0.09	0.21					
Pb	6	15	19	10	8	6	16	13	12	10	7	9	9					
Th	4.46	5.49	3.89	4.09	5.01	3.88	4.97	4.98	4.61	4.4	4.43	3.54	4.38					
U	1.59	1.57	2.53	1.79	1.86	1.41	2	1.58	1.21	1.15	1.15	1.1	1.2					
K-Ar age	11.27	12.52	12.74	13.2	11.4	11.56	11.64	11.22	12.21	11.55	11.31	11.28	10.75	13.39	13.49	13.36	12.66	11.62

LOI = total volatile lost on ignition. Mg# = (Mg/(Mg+Fe^{2+}))×100, with Fe^{3+}/Fe^{2+} as suggested by Middlemost (1989)