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Pre-Alpine evolution of a segment of the North-Gondwanan margin: Geochronological and geochemical evidence from the central Serbo-Macedonian Massif

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ABSTRACT

The Serbo-Macedonian Massif (SMM) represents a composite crystalline belt within the Eastern European Alpine orogen, outcropping from the Pannonian basin in the north, to the Aegean Sea in the south. The central parts of the massif (i.e. southeastern Serbia, southwestern Bulgaria, eastern Macedonia) consist of the medium- to high-grade Lower Complex, and the low-grade Vlasina Unit. New results of U–Pb LA-ICP-MS analyses, coupled with geochemical analyses of Hf isotopes on magmatic and detrital zircons, and main and trace element concentrations in whole-rock samples suggest that the central SMM and the basement of the adjacent units (i.e. Eastern Veles series and Struma Unit) originated in the central parts of the northern margin of Gondwana. These data provided a basis for a revised tectonic model of the evolution of the SMM from the late Ediacaran to the Early Triassic.

The earliest magmatism in the Lower Complex, Vlasina Unit and the basement of Struma Unit is related to the activity along the late Cadomian magmatic arc (562–522 Ma). Subsequent stage of early Palaeozoic igneous activity is associated with the reactivation of subduction below the Lower Complex and the Eastern Veles series during the Early Ordovician (490–478 Ma), emplacement of mafic dykes in the Lower Complex due to aborted rifting in the Middle Ordovician (472–456 Ma), and felsic within-plate magmatism in the early Silurian (439 ± 2 Ma). The third magmatic stage is represented by Carboniferous late to post-collisional granites (328–304 Ma). These granites intrude the gneisses of the Lower Complex, in which the youngest deformed igneous rocks are of early Silurian age, thus constraining the high-strain deformation and peak metamorphism to the Variscan orogeny. The Permian–Triassic (255–253 Ma) stage of late- to post-collisional and within-plate felsic magmatism is related to the opening of the Mesozoic Tethys.

1. Introduction

The Alpine orogen is probably the best studied orogenic belt and yet there are several parts where the geological history is still very poorly resolved. These inadequately studied areas hinder the accuracy of global tectonic models, especially those concerning the pre-Alpine evolution. Prime example of such areas are the crystalline basement units of southeastern Europe, which were often omitted from the large-scale tectonic models. However, the last twenty years brought significant amount of knowledge concerning the palaeogeographic position and tectonic evolution of a number of crystalline units previously considered to be ancient microcontinents trapped within the Eastern European Alpine orogenic belt (e.g. Tisza, Rhodope; Kober, 1921). These recent studies revealed that such terranes represent complex collages of reworked continental (and locally oceanic) crust and sediments actively involved in several phases of Alpine deformation and metamorphism (e.g. Ivanov, 1988; Burg et al., 1990, 1996; Liati and Gebauer, 1999; Burg, 2012). Among these terranes, the tectonic position and evolution of the Serbo-Macedonian Massif (SMM) outcropping in Serbia, southwest Bulgaria, Macedonia, and northern Greece, remains enigmatic (Fig. 1). It becomes crucial for valid reconstruction of the long and complex interaction of Gondwana- and Laurussia-derived crustal segments presently outcropping along the Balkan Peninsula (e.g. Munteanu and Tatu, 2003; Himmerkus et al., 2009a; Kalvoda and Bábek, 2010; Meinhold et al., 2010; Kroner and Romer, 2013), to resolve the enigma concerning the provenance and geological history of the Serbo-Macedonian Massif. Recent studies in the Greek part of the SMM reveal a complex

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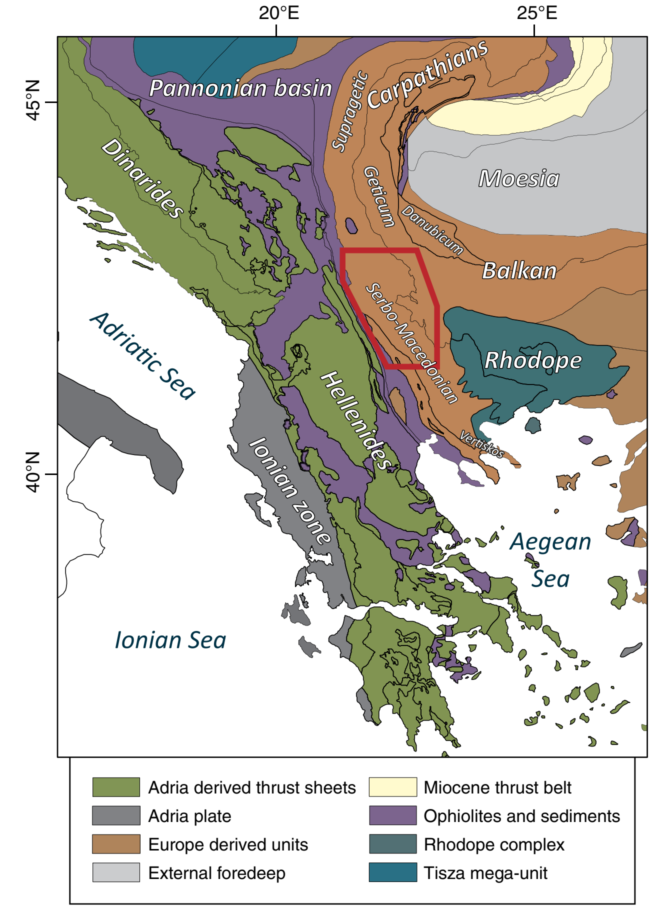


Fig. 1. Tectonic sketch of the Balkan Peninsula (after Schmid et al., 2008; van Hinsbergen and Schmid, 2012) with the position of the study area (dark polygon).

metamorphic and tectonic history since the Cambrian, including intense Alpine overprint suggesting that what we today refer to as the Serbo-Macedonian Massif is not a single tectonic unit (e.g. Kilias et al., 1997; Brun and Sokoutis, 2007; Himmerkus et al., 2007).

In order to provide a basis for reliable reconstruction of the early tectonic history of crystalline units in southeastern Europe, this article presents evidence for the origin and evolution of the SMM obtained through analyses of U–Pb isotopic ages of magmatic and detrital zircons coupled with Hf isotope analysis of the dated zircons and whole-rock geochemical analysis of major and trace elements. The analyses were performed on a wide range of rock types in all units constituting the SMM in southeastern Serbia, southwestern Bulgaria and eastern Macedonia, with additional samples from the adjacent tectonic domains occasionally reported as parts of the SMM (Brković et al., 1980; Dolić et al., 1981; Karamata and Krstić, 1996; Dimitrijević, 1997). The results of the geochronological and geochemical analyses allow correlation with similar units of the Eastern Mediterranean Alpine orogen. These data are used to propose a model of tectonic evolution of the study area and adjacent domains since the late Neoproterozoic until the earliest Mesozoic. This dataset will serve as a sound basis for further research including more detailed sedimentary provenance analyses and geochemical correlations in the wider region, which could correct the deficiencies in the palaeogeographic reconstructions and tectonic models concerning the early evolution of tectonic units presently located in the Eastern Mediterranean Alpine orogen.

2. Geological setting

The Serbo-Macedonian Massif extends from the Aegean Sea to the Southern Carpathians (Fig. 1) where it is correlated with the Supragetic nappe sequence (Dimitrijević, 1997; Iancu et al., 2005). It should be noted that although our study concerns only the pre-Alpine evolution of the SMM, the names of the tectonic units discussed below refer to the Alpine orogen so that results presented here could be correlated with the previously published tectonic frameworks of southeastern Europe. The SMM in Serbia and Macedonia represents an entirely metamorphic belt comprising a structurally lower unit (the Lower Complex) and an upper unit (Vlasina Unit), as originally proposed by Dimitrijević (1957). These units are commonly differentiated on the basis of their metamorphic grade, with the Lower Complex predominantly metamorphosed at medium to lower amphibolite facies, and the Vlasina Unit at greenschist facies. The boundary of these two units is usually reported as tectonic, i.e. the Vrvi Kobila shear zone (Fig. 2). This contact was previously described as a pre-Mesozoic west-vergent brittle thrust of Vlasina Unit over the Lower Complex (Vukanović et al., 1973; Krstić and Karamata, 1992), or a post-Late Cretaceous dextral shear zone (Kräutner and Krstić, 2002). The continuations of the Lower Complex and Vlasina Unit in Bulgaria are referred to as the Ograzhden block (Dimitrijević, 1967; Zagorchev, 1984a; Dabovski et al., 2002) and Morava Unit (Zagorchev, 1985; Zagortchev, 1993), respectively. The Vertiskos Unit in Greece is traditionally considered as the continuation of the Lower Complex of the SMM (Kockel et al., 1971; Burg et al., 1995; Himmerkus et al., 2009a; Meinhold et al., 2010; Burg, 2012). Based on the available geological records from the Greek and Bulgarian extents of the SMM, its provenance was previously assigned to the eastern Avalonian (Meinhold et al., 2010) or Cadomian (Stampfli et al., 2002; von Raumer et al., 2003; Balintoni et al., 2010b; Meinhold et al., 2010; Kounov et al., 2012) assemblages of terranes within the Neoproterozoic north-Gondwanan arc.

During the late Early Cretaceous, the Vlasina Unit was thrust to the east onto the Getic units along a system of east- to northeast-vergent thrusts (Petković, 1930; Mihailescu et al., 1967; Petrović, 1969; Zagorchev and Ruseva, 1982; Zagorchev, 1984b; Lilov and Zagorchev, 1993; Kounov et al., 2010). In southwestern Bulgaria and northern Greece the contact of the Ograzhden block and the Vertiskos Unit with the Rhodope complex is traced along the Strymon Valley and Kerdillion detachments (Dinter and Royden, 1993; Brun and Sokoutis, 2007; Kounov et al., 2015). The distinction between the SMM and the Rhodope complex was often challenged in the past (Popović, 1991; Ricou et al., 1998; Grubić et al., 1999, 2005), although their differences were firmly established in the Greek and Bulgarian parts of the SMM during the past two decades (Burg, 2012 and references therein). A large number of studies in Bulgaria and Greece have shown that the Rhodope complex was formed during the Alpine orogeny (e.g. Burg et al., 1996; Kaiser-Rohrmeier et al., 2004; Liati, 2005; Bosse et al., 2009; Turpaud and Reischmann, 2010), whereas the SMM lacks the high-grade Cenozoic overprint (Georgiev et al., 2010; Kounov et al., 2010, 2012).

The SMM is bounded to the west by Mesozoic sediments and Jurassic ophiolites of the Eastern Vardar suture zone (e.g. Karamata, 2006; Schmid et al., 2008; Meinhold et al., 2009; Robertson et al., 2009). In the study area this contact is reported as a strike-slip fault, an east-vergent thrust (Krstić and Karamata, 1992), or a westward thrust which was partially reactivated as a dextral strike slip fault in the Neogene (Kräutner and Krstić, 2002). South of the Lece andesitic complex, the Lower Complex of the SMM is in tectonic contact with the metamorphic rocks of the Eastern Veles series (Fig. 2). The oldest reported, non-metamorphosed, sedimentary rocks that overlie crystalline rocks of the SMM are Middle Triassic (Karamata and Krstić, 1996; Dimitrijević, 1997).

Earlier studies suggest that being previously variably metamorphosed, the Lower Complex and Vlasina Unit have been amalgamated and subsequently accreted to the Moesian platform during the Variscan orogeny (Karamata and Krstić, 1996; Haydoutov and Yanev, 1997). An alternative scenario proposes that the southern extent of the SMM (i.e. Vertiskos Unit) had joined the European craton in the Late Jurassic to Early Cretaceous (Himmerkus et al., 2009a). Schmid et al. (2008) suggest that the Eastern Vardar ophiolites are at a structurally higher position in respect to the SMM due to the Late Jurassic obduction and final

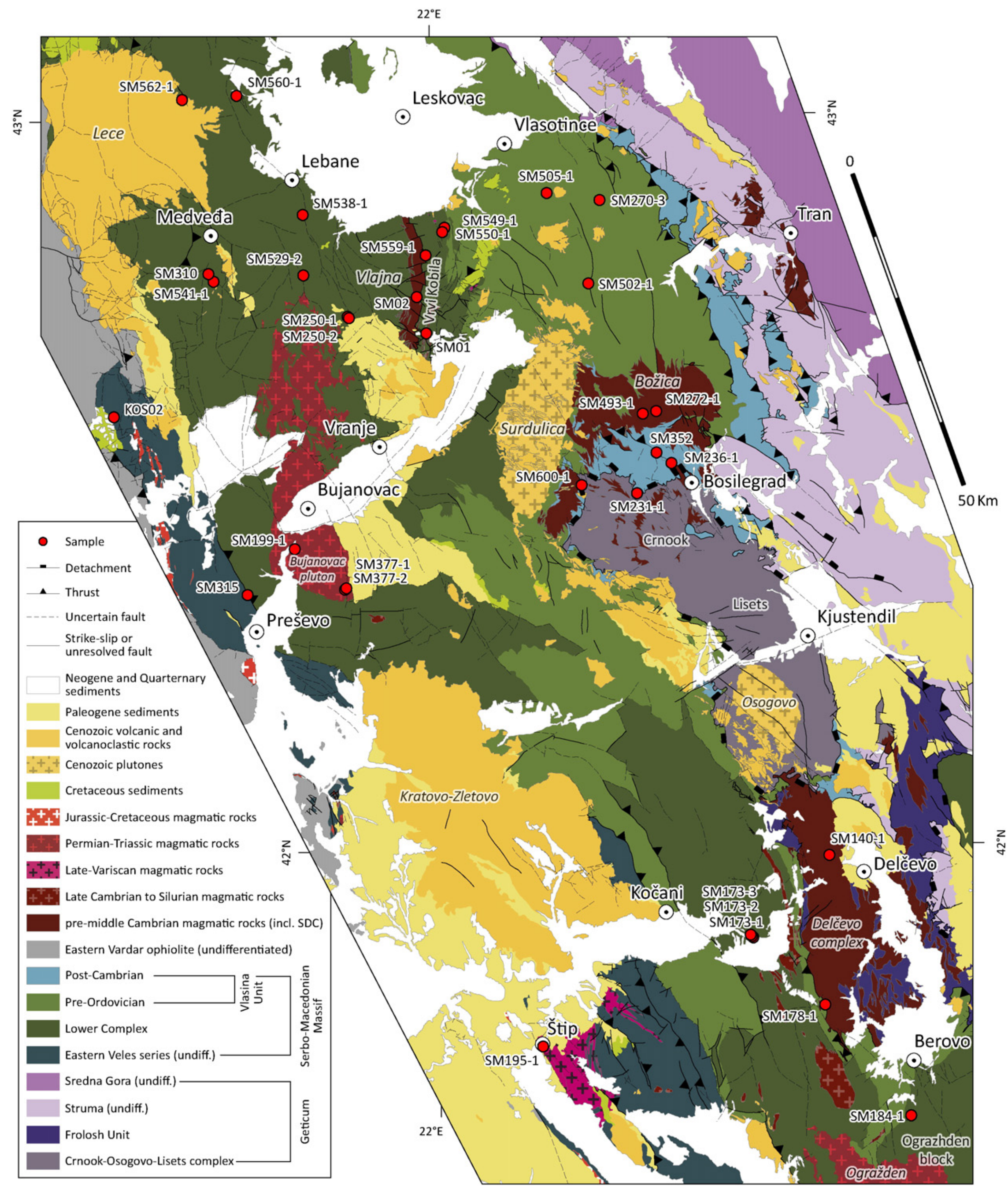


Fig. 2. Tectonic map of the study area with the position of the analysed samples (after Basic geological maps of SFR Yugoslavia 1:100 0000 and Geological maps of Bulgaria 1:100 000 and 1:50 000). Names in italic represent magmatic bodies.

east-facing nappe stacking in the Late Cretaceous. According to these authors the final suturing in the early Palaeogene, related to the final closure of the remnants of the Vardar Ocean (i.e. the Sava zone), led to the westward thrusting in the Dinarides (Adria-derived).

2.1. The Lower Complex

The Lower Complex consists mainly of gneisses, micaschists, quartzites with lenses of amphibolites and occasionally marbles and migmatites (Dimitrijević, 1963, 1997). Isolated fragments of serpentinised ultramafic rocks were also reported (Petrović et al., 1973; Vukanović et al., 1973; Nenova and Marinova, 2007; Tarassov et al., 2007). Pelitic and psammitic sediments are usually considered as protoliths of the felsic metamorphic rocks, and tholeiitic within-plate basalts and related tuffs as protoliths of the amphibolites (Milovanović, 1992; Dimitrijević, 1997). The protoliths of the Lower Complex and Vlasina Unit are described as parts of an arc-related volcano-sedimentary series (Dimitrijević, 1967; Krstić and Karamata, 1992; Haydoutov and Yanev, 1997; Kounov et al., 2012). Although a medium amphibolitic facies metamorphism was suggested for the Lower Complex and the Ograzhden block (Milovanović, 1990; Cvetković et al., 1995; Zidarov et al., 2003a; Nenova and Zidarov, 2008; Erić et al., 2009), relict kyanite and boudins of eclogites were also reported

(Balogh et al., 1994; Fed'kin et al., 1996; Vasković and Tasić, 1997; Iancu et al., 1998; Korikovsky et al., 2003; Nenova and Zidarov, 2008). This high pressure event was related to the late Neoproterozoic or Early Palaeozoic (Balogh et al., 1994; Zagorchev and Milovanović, 2006; Nenova and Zidarov, 2008), while the amphibolite facies overprint is assigned to the Variscan orogeny (Dimitrijević, 1967; Medaris et al., 2003; Nenova and Zidarov, 2008). A latest Jurassic and/or Early Cretaceous greenschist facies retrogressive stage was reported in Serbia, Bulgaria and Greece (Milovanović, 1990; Balogh et al., 1994; Kilias et al., 1999; Zidarov et al., 2003a).

West of the Vrvi Kobila shear zone, the Lower Complex is intruded by the Vlajna granitoid (Fig. 2), which was reported to have been emplaced at 450 Ma (Dimitrijević, 1963). The largest magmatic body intruding the Lower Complex in Serbia is the north to northeast-south to southwest elongated Bujanovac S-type granite (Vukanović et al., 1973, 1977; Karamata and Krstić, 1996). Two distinct intrusions form the Bujanovac granitic complex, an older represented by medium-grained foliated granitoids, intruded by younger, fine-grained granites (Dimitrijević, 1958; Vukanović et al., 1977). The ages of these plutons have been reported as 347 Ma (Dimitrijević, 1997), and 234 Ma (Dimitrijević, 1958), respectively.

A group of magmatic bodies, apparently intruding the Vlajna granites, located in the southern part of the Vrvi Kobila shear zone are referred to as Kukavica granites (Fig. 2). Both Vlajna and Kukavica granites were deformed together along the Vrvi Kobila shear zone.

The Ogražden pluton intruded into the Lower Complex (i.e. Ograzhden block) gneisses on the southern margin of the study area (Fig. 2), consists of biotitic granites and granodiorites. Age of its emplacement is reported as early Palaeozoic (Boev et al., 2002), Early Cretaceous (Rakićević et al., 1973), and most recently as Early Triassic (252 ± 2 Ma; Georgiev et al., 2012).

2.2. Vlasina Unit

Karamata and Krstić (1996) regarded Vlasina Unit as a part of the Suprageticum (i.e. highest structural unit in the Carpatho-Balkanide orogen), due to the suggested hanging wall position in the thrust over the Lower Complex (i.e. Vrvi Kobila shear zone, Fig. 2). Additionally, several reports suggest that these units originally shared a stratigraphic contact, which was later tectonically reactivated (Petrović and Karamata, 1965; Dimitrijević, 1997).

The Vlasina Unit consists of the pre-Ordovician greenschist-facies basement rocks, overlain by a low-grade post-Cambrian sedimentary sequence (Kräutner and Krstić, 2002). Both parts show a similar style of deformation and metamorphism exhibiting a consistent gradient throughout the area. The apparent abrupt changes in the metamorphic grade observed at certain localities could be explained by subsequent telescoping due to brittle thrusting.

Although regarded as a volcano-sedimentary complex similarly as the Lower Complex, the rocks of the pre-Ordovician Vlasina are dominated by ocean-floor sediments and contain basic magmatic rocks related to MORB (Popović, 1991), representing an ocean-ward periphery of a magmatic arc (Milovanović et al., 1988). The Pre-Ordovician Vlasina is represented by chlorite, biotite, muscovite, sericite and epidote schists. Local varieties rich in quartz and albite, or actinolite and stilpnomelane were occasionally reported (Dimitrijević, 1967). Phyllites and quartzites are also common. Protoliths of these metamorphic rocks are pelitic and to a lesser extent psammitic sediments (Petrović, 1969), together with arc-related tholeiitic basalts and their tuffs (Milovanović et al., 1988). Several small, tectonically imbricated bodies of serpentinites have been reported (Ilić et al., 1967; Petrović, 1969; Petrović et al., 1973). Intrusions of granites, gabbro and diabase are often deformed and metamorphosed along with the country rock. The age of the pre-Ordovician Vlasina protoliths was determined as "Ripheo-Cambrian" based on the spores recovered at several locations in southeastern Serbia (Pantić et al., 1967; Petrović, 1969). Additionally, lenses of calcschists northeast of Surdulica pluton yielded palynomorphs and fungi of Cambrian age (Vasković and Tasić, 1997 and references therein).

At the type locality of Vlasina Unit (northeast of Surdulica pluton near sampling location of SM502-1, Fig. 2) the peak metamorphic conditions reached greenschist facies (Milovanović et al., 1988; Vasković, 2002). However, relict amphibolite and transitional amphibolite-greenschist facies assemblages were reported in the Vrvi Kobila area (Petrović, 1969), vicinity of the Božica magmatic complex (Babović et al., 1977; Krstić and Karamata, 1992), north of the Surdulica pluton (Pavlović, 1977) and east of Vranje (Babović et al., 1977; Vasković, 1998; Vasković et al., 2003). This higher grade metamorphism in the Vlasina Unit has been associated with the "Baikalian" (850–650 Ma) and "Caledonian" orogenic events (Karamata and Krstić, 1996), and the age of the greenschist overprint as early Carboniferous (Dimitrijević, 1963; Petrović, 1969; Krstić and Karamata, 1992; Karamata and Krstić, 1996; Graf, 2001), or post-Variscan (Dimitrijević, 1967).

The pre-Ordovician Vlasina is covered by a meta-sedimentary succession reported as Ordovician to lowest Carboniferous in age (Spassov, 1973; Babović et al., 1977; Pavlović, 1977; Lakova, 1997). These meta-sedimentary rocks crop out along the eastern margin of the Vlasina Unit, often separated from the pre-Ordovician Vlasina by east-vergent thrusts (Petrović, 1969; Fig. 2). The originally transgressive contact with the pre-Ordovician Vlasina reported by Pavlović (1962, 1977) northeast of Bosilegrad, is in fact obscured by intensive deformational overprint. The lowermost part of the sedimentary column comprises phyllites, metasandstones, calcschists, quartzite, marble, sericite and chlorite schists (Babović et al., 1977). Inarticulate brachiopod fauna in the "basal quartzites" suggests an Early Ordovician age (Pavlović, 1962). The Ordovician period is represented upward by argillo-phylites, spilites, meta-pelites, quartz and sericitic schists with lenses of quartzite and calcschist (Petrović et al., 1973). The Silurian succession continues with graptolitic schists, conspicuous carbonates with Middle Devonian spillites, ending with turbidites of Upper Devonian to lowest Carboniferous age (Petrović, 1969; Petrović et al., 1973; Spassov, 1973; Krstić, 1981; Krstić et al., 2002; Banjac, 2004; Lakova, 2009; Boncheva et al., 2010).

A large complex of magmatic rocks located east of Surdulica (Fig. 2), which was intruded into the pre-Ordovician Vlasina is referred to as Božica granitoid in Serbia (Petrović et al., 1973; Babović et al., 1977), and Milevski granite in Bulgaria (e.g. Graf, 2001; Kounov et al., 2012). It comprises partially deformed granites, diorites and gabbros. A late Cadomian age (551 ± 1 Ma) was determined in the Bulgarian part of this magmatic complex (Kounov et al., 2012), whereas an age of 500 Ma was suggested for the Serbian part of Božica (Kräutner and Krstić, 2002).

2.3. Geticum

The Geticum represents a large-scale tectonic zone in the Alpine tectonic framework of the Carpatho-Balkan orogen, comprising a number of tectonic units forming southeast-, east- and northeast-vergent nappes (e.g. Struma, Sredna Gora). The crystalline basement of the Struma Unit consists of variably deformed continent- and ocean-derived rocks of Ediacaran to early Cambrian protolith age (e.g. Kounov et al., 2012), unconformably overlain by a Permian to Lower Cretaceous sedimentary cover (Zagorchev, 1981). The ophiolites (Frolosh Unit) and a magmatic-arc igneous suite, the Struma Diorite Complex (SDC; Stephanov and Dimitrov, 1936; Haydoutov et al., 1994) are tectonically imbricated (Fig. 2). Although the present-day contacts between the fragments of ophiolitic ultramafic rocks, gabbros, diabases, basic tuffs and minor clastic metasedimentary rocks of the Frolosh Unit, and the gabbros, gabbrodiorites, diorites and granites of SDC are mainly tectonic, primary intrusive contacts can be locally observed. The mafic rocks of the Frolosh ophiolites show affinity to MORB (Haydoutov and Pin, 1993; Kounov et al., 2012). The rocks of the Lower Complex (Ograzhden block; Zagorchev, 1984a), situated in the southeast of the study area

(Fig. 2), are separated from the overlying SDC and Frolosh Unit by the Gabrov Dol detachment east of the study area (Bonev et al., 1995).

The Crnook–Osogovo–Lisets complex (COL; Fig. 2), consists of amphibolites, micaschists, muscovite-biotite and amphibole-biotite gneisses (Dimitrova, 1964; Antić et al., 2014). Lenses of ultramafic rocks were also reported (Haydoutov et al., 1994). The magmatic rocks of the SDC and the Crnook–Osogovo–Lisets complex are derived from the same calc-alkaline magma source and represent a magmatic-arc formed during the Ediacaran to early Cambrian (Kounov et al., 2012). The lower amphibolite-facies metamorphism and deformation in the COL complex were associated with late Early Cretaceous compression in the area (Kounov et al., 2010). The COL metamorphic rocks were exhumed from below the SDC during the middle Eocene–Oligocene extension (Kounov et al., 2004, 2010; Antić et al., 2014).

2.4. Eastern Veles series

South of the Lece volcanic complex (Fig. 2), the Lower Complex is in tectonic contact with the metamorphic series of amphibolites, micaschists, gneisses, quartzites and marbles (Fig. 2). These rocks are regarded as part of the Veles series (Karajovanović and Hristov, 1976; Vukanović et al., 1977), which is considered as part of the Circum-Rhodope belt (definitions provided in Zagorchev and Milovanović, 2006; Kounov et al., 2011; Schmid et al., 2014), or internal Vardar zone (Dimitrijević and Drakulić, 1958; Dimitrijević, 1997; Robertson et al., 2009). The Veles series is separated in the eastern and the western part by Jurassic ophiolites and Upper Cretaceous sediments (Vukanović et al., 1977; Pavić et al., 1983; Karamata and Krstić, 1996). The Lower Complex in the study area is in direct contact only with the latter (Fig. 2). The Eastern Veles series comprises Cambrian to Devonian sediments (Pavlović, 1977), initially metamorphosed under amphibolite facies conditions, preserved only as relicts due to intensive greenschist retrogression (Dimitrijević, 1997). The crystalline rocks of the Eastern Veles series are covered by Triassic sediments metamorphosed to greenschist facies, present only in a small area in the northwestern periphery of the unit (Pavić et al., 1983). In contrast, the Western Veles series comprises predominantly Upper Devonian to Triassic metasedimentary rocks (Pavić et al., 1983; Grubić and Ercegovac, 2002), and low-grade igneous rocks related to a Carboniferous magmatic arc (Karamata, 2006). Most previous authors agree that the crystalline rocks of the Eastern Veles series (excluding the Triassic cover) were originally a part of the Lower Complex which have subsequently suffered intensive Alpine overprint, less pronounced in the rest of the Lower Complex (Brković et al., 1980; Dolić et al., 1981; Karamata and Krstić, 1996; Dimitrijević, 1997). Another distinctive feature of the Eastern Veles series compared to the Lower Complex of the SMM is the higher occurrence of marble (Dimitrijević, 1997).

The voluminous Štip magmatic complex represents a heterogeneous magmatic body comprising granites, monzonites and granodiorites, which intruded the Eastern Veles series near the town of Štip (Fig. 2). They contain xenoliths of serpentinite and amphibolite schists, which were considered as Jurassic by Rakićević et al. (1976), and Palaeozoic by Karajovanović and Hadži-Mitrova (1982). The upper limit on the age of emplacement of the Štip complex is constrained by the Albian–Cenomanian (i.e. 113–94 Ma) sediments covering the southeastern periphery of the pluton. Rb–Sr dating of biotite from the Štip complex yielded an age of 161 ± 3 Ma which was interpreted as the age of its emplacement (Šoptrajanova, 1967), or as a metamorphic overprint (Spray et al., 1984).

3. Samples and analytical methods

Sample descriptions with locations are presented in Table 1. Twenty two samples were taken from the Lower Complex, eight from the Vlasina Unit, three from the Eastern Veles series, and two samples were taken from the COL complex. Major element analyses on whole-rock samples were performed using the X-ray fluorescence (XRF) method, whereas the trace element and REE measurements were carried out with Laser Ablation Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS) at ETH Zürich. All diagrams representing results of geochemical analyses were plotted using GCDkit software (Janoušek et al., 2006). The results of the chemical analyses are presented in Appendix 1. Whole rock samples were mechanically fragmented (jaw crusher, milling) and sieved through a 450 μm sieve. A heavy-minerals concentrates were obtained by gravity separation (Wilfley table). Zircon concentrates were recovered by conventional heavy liquid (methylene iodide 3.3 $g \cdot cm^{-3}$) and magnetic susceptibility (Frantz isodynamic separator) separation methods. Extracted zircon grains were hand-picked and mounted in epoxy resin. The polished mounts were photographed, and SEM cathodoluminescence (CL) and backscatter electron images were taken in order to analyse the internal structure of the grains prior to the LA-ICP-MS analyses. Instrument parameters used during the course of this study are presented in detail in Appendix 2. First round of data acquisition was performed at Department of Earth Science, ETH Zürich in 2010 using an Elan 6100 ICP-MS (PerkinElmer, Norwalk, CT, USA) coupled to an 193 nm ArF-Excimer laser ablation system similar to a Geolas system (Coherent, USA). The laser was operated at 10 Hz, spot size was 40 μm and a fluence of 4.0 $J \cdot cm^{-2}$ was used. Data obtained by this set-up are designated with an “o” as a prefix to the spot name. The second round of measurements were made in 2012 (ETH Zürich), using an Element-XR SF-ICP-MS (Thermo Fisher, Bremen, Germany) coupled with an 193 nm Excimer laser (Resonetics Resolution S155-LR) that was operated at 5 Hz and a fluence of 2.0 $J \cdot cm^{-2}$. The spot size of 30 μm was used to obtain the data in this round of measurements. All experiments were performed using helium as carrier gas. The carrier gas was mixed with argon as make-up gas before entering the ICP (Appendix 2).

Appendix 3 contains detailed descriptions of samples and obtained U–Pb ages, a comprehensive table with results of the U, Th and Pb isotopic measurements is presented in Appendix 4, while the CL images of analysed zircon grains are provided in Appendix 5. Unless stated otherwise, concordia diagrams and density distribution plots were produced using the programs Isoplot (v3.71.09.05.23nx, Ludwig, 2012) and DensityPlotter (v.6.1; Vermeesch, 2012), respectively. Unless explicitly stated, uncertainties in the calculated weighted mean ages are reported at 95% confidence limit. Results referred to as concordant are within 95–105% tolerance defined by $100 \cdot ((^{206}Pb/^{238}U \text{ age}) \cdot (^{207}Pb/^{235}U \text{ age})^{-1})$. Weighted average plots do not include discordant measurements. Since annealing and chemical leaching were not performed on the analysed zircon grains, the results obtained on partially metamict domains might yield concordant ages that can be possibly younger than the real age due to partial radiogenic lead loss. In cases where a group of age results forms a continuous array lacking “plateau” or evident clustering related to specific growth domains, the weighted average of the array is reported as the most correct result. Discordant measurements and results from spots located in the “mixed zones” within the zircon grain (e.g. on the interface of core and rim), were omitted from the interpretation and the plots. However, discordant results are plotted on concordia diagrams in Appendix 3.

4. Results

4.1. Geochemistry

4.1.1. The Lower Complex

Major oxide analysis of eleven sampled felsic magmatic rocks and gneisses from the Lower Complex (Appendix 1) indicates predominant calc-alkaline granitic composition (Fig. 3a), with a subordinate intermediate composition of the Vlajna granitoid (SM02; Fig. 2), deformed Bujanovac Qz-monzonite (SM377-2) and Vinica gneiss (SM173-2 in Fig. 3a). Amphibolites from the Vinica area east of Kočani (SM173-1; Fig. 2) and Dobra Voda locality east of Lece (SM562-1; Fig. 2) are

Table 1
Sample descriptions and locations.

Sample	Rock type	Gauß-Krüger[1]		DMS[2]		Locality	Analyses[3]
		x	y	x	y		
Lower Complex							
SM01	Mylonite	7581020	4729839	21°59′3.29″E	42°42′12.496″N	Ovča Strana	3
SM02	Granodiorite	7579581	4735334	21°58′1.844″E	42°45′8.702″N	Kukavica	3
SM173-1	Amp-schist	7630610	4638229	22°34′25.481″E	41°52′36.782″N	Vinica	3
SM173-2	Two-mica gneiss	7630610	4638229	22°34′25.481″E	41°52′36.782″N	Vinica	1
SM173-3	Leucocratic dyke	7630610	4638229	22°34′25.481″E	41°52′36.782″N	Vinica	3
SM184-1	Two-mica gneiss	7654525	4611248	22°50′58.068″E	41°37′27.378″N	Maleševski Mts.	3
SM199-1	Granite	7561075	4697110	21°43′40.285″E	42°24′12.474″N	Borovac	3
SM250-1	Amphibolite	7569098	4732307	21°50′12.623″E	42°43′32.618″N	Golemo Selo	3
SM250-2	Paragneiss	7569098	4732307	21°50′12.623″E	42°43′32.618″N	Golemo Selo	2
SM310	Paragneiss	7548389	4738347	21°35′6.753″E	42°47′1.236″N	Sijarinska Banja	2
SM377-1	Weakly deformed granite	7568705	4690941	21°50′2.944″E	42°21′33.452″N	Spančevac	3
SM377-2	Deformed Qz-monzonite	7568705	4690941	21°50′2.944″E	42°21′33.452″N	Spančevac	3
SM529-2	Amphibolite	7562402	4738588	21°45′40.644″E	42°47′11.034″N	Šumanska Reka	1
SM538-1	Metagabbro	7562291	4747753	21°44′26.448″E	42°51′40.215″N	Popovce	1
SM541-1	Amphibolite	7548798	4737626	21°35′30.155″E	42°46′39.822″N	Sijarina	1
SM549-1	Metagabbro	7583799	4745848	22°1′23.325″E	42°51′2.239″N	Mala Oraovica	1
SM550-1	Granite	7583796	4745423	22°1′23.855″E	42°50′40.234″N	Slatinska Reka	2
SM559-1	Granite	7580913	4741665	21°59′22.368″E	42°48′53.175″N	Muratovica	1
SM560-1	Metagranite	7552276	4765813	21°38′8.152″E	43°1′51.78″N	Brestovac	1
SM562-1	Amphibolite	7543986	4765206	21°31′53.954″E	43°1′25.933″N	Dobra Voda	1
Vlasina Unit							
SM178-1	Ep-Chl-Amp schists	7641503	4628070	22°41′53.009″E	41°46′45.925″N	Mitrašinci	1
SM270-3	Diabase	7607295	4750047	22°18′49.385″E	42°53′13.117″N	Dobro Polje	1
SM272-1	Granite	7615854	4718033	22°24′17.378″E	42°35′33.171″N	Božica	3
SM352	Gabbro	7615948	4711742	22°24′33.416″E	42°32′21.373″N	Donja Lisina	3
SM493-1	Diorite	7613838	4717645	22°22′50.226″E	42°35′17.209″N	Toplodolska Reka	1
SM502-1	Komatiite	7605584	4737380	22°16′50.478″E	42°46′3.893″N	Veljkovci	1
SM505-1	Ab-gneiss	7599255	4751127	22°12′23.016″E	42°53′35.606″N	Gornja Lopušnja	1
SM600-1	Metagranite	7604563	4706769	22°15′19.318″E	42°29′29.268″N	Gornja Ljubata	2
Eastern Veles series							
KOS02	Micaschist	7533668	4717085	21°24′33.357″E	42°35′43.487″N	Izvor	2
SM195-1	Bt-granite	7598733	4621701	22°11′0.429″E	41°43′44.482″N	Štip	3
SM315	Orthogneiss	7553957	4690113	21°38′54.581″E	42°21′1.032″N	Bukovac	3
Basement of Struma Unit							
SM140-1	Granite	7642147	4650734	22°42′42.195″E	41°58′59.254″N	Delčevo	3
SM231-1	Gabbro	7612988	4705575	22°22′19.907″E	42°28′56.439″N	Donja Ljubata	1
SM236-1	Monzonite	7618183	4710189	22°26′17.931″E	42°31′25.908″N	Bosilegrad	3

[1] Gauß-Krüger coordinate system is used in SFR-Yugoslavian Basic Geological 1:100,000 maps.
[2] Coordinates given in degree-minute-second (DMS) format.
[3] Explanation of values: 1 — geochemical analysis only; 2 — U–Pb age determination only; and 3 — geochemical analysis and U–Pb age determination.

presented in this section as they are classified as intermediate magmatic rocks (i.e. gabbro diorites in Fig. 3a). These three samples represent the tholeiitic type of rocks in this group (Fig. 3b).

The majority of granitic rocks from the Lower Complex plot within the volcanic-arc granites field (Fig. 4). The sample SM02 from the southern part of the Vlajna granitoid and a leucocratic dyke from Vinica area (SM173-3, east of Kočani, Fig. 2) are slightly enriched in Y and Nb compared to the remaining arc-related rocks of the Lower Complex. This Y enrichment coupled with their peraluminous signature (Fig. 5) reveals involvement of crustal material in their magmatic source. The peraluminous Kukavica granite in the Vrvi Kobila area (SM01, Figs. 4 and 6) plots closer to the field of syn-collisional granites probably due to enrichment in Rb. High contents of Ta in fine-grained granites from the periphery of Bujanovac pluton (SM377-1) and Maleševski Mts. orthogneiss (SM184-1) suggest within-plate setting for these rocks (Fig. 6a).

Most of the analysed acidic and intermediate rocks from the Lower Complex show enrichment in light rare earth elements (LREE) (Fig. 7a left). The samples from the Bujanovac magmatic complex (SM199-1, SM377-1 and SM377-2) as well as central Vlajna granite (SM559-1), Kukavica granite (SM01) and gneiss sample SM184-1 show slight depletion in heavy rare earth elements (HREE). The distinct negative Eu anomalies detected in the Vinica leucocratic dyke (SM173-3), the Kukavica (SM01), central Bujanovac (SM199-1) and central Vlajna (SM559-1) granites, as well as Maleševski Mts. orthogneiss (SM184-1) could be attributed to crystal fractionation of plagioclase.

Trace element diagram, normalised to primitive mantle (Hofmann, 1988), reveals general affinity to magmatic arc environment of most of the analysed samples (grey field in Fig. 7a right). This is inferred from their enrichment in incompatible elements (i.e. Rb, Ba, Th and U) and relative depletion of Nb and Ta. The exceptions include samples SM02, SM562-1 and SM559-1 that display depletion in Sr or higher concentration of HREE indicative of their affinity to within-plate setting. Furthermore, the coarse-grained Bujanovac Qz-monzonite (SM377-2) is relatively enriched in Ba, Nb, Ta, Hf and Zr supporting within-plate origin despite slightly elevated Sr and lower HREE concentrations (Fig. 7a right).

Amphibolites and meta-gabbros from the Lower Complex (Appendix 1), chemically equivalent to gabbros (Fig. 3a), are associated with tholeiitic series (Fig. 3b). Most of the analysed samples are designated as enriched MORB or within-plate tholeiites (Fig. 6b) except Sijarina amphibolites (SM541-1) that plot as within-plate alkali-basalt, and Slatina meta-gabbro (SM549-1) pertaining to the continental-arc environment. Most of the sampled basic rocks from the Lower Complex display a relatively enriched pattern in REE with slight downward sloping towards HREE (Fig. 7b left). Exceptions are Sijarina amphibolites (SM541-1, south of Medveđa, Fig. 2) with more pronounced depletion of HREE, and Gornji Brestovac meta-gabbro (SM560-1, north to

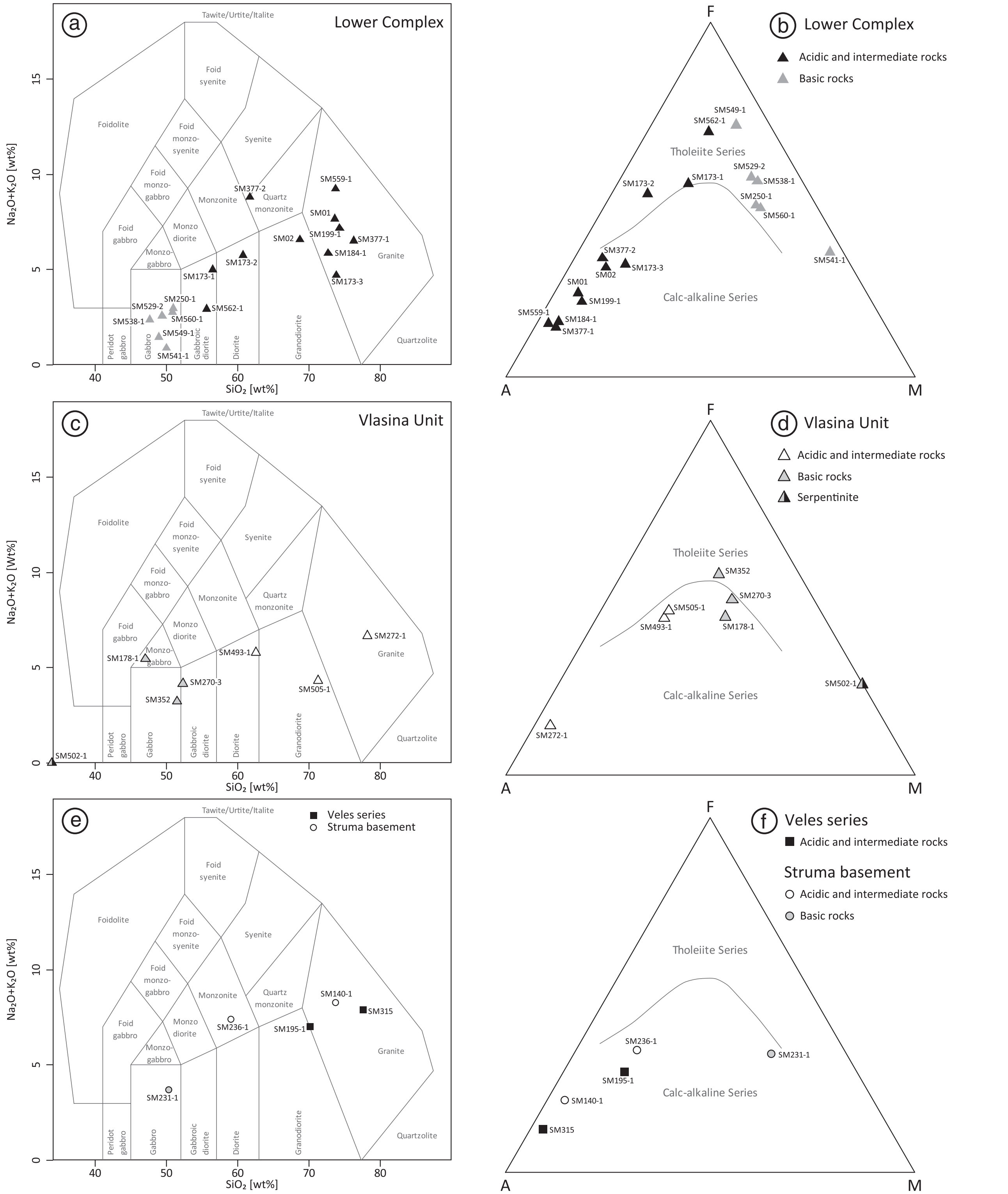


Fig. 3. (a), (c) and (e) Total alkali versus silica classification plots (Middlemost, 1985); (b), (d) and (f) AFM discrimination plots (Irvine and Baragar, 1971) of all analysed samples.

northeast of Lebane, Fig. 2) that shows excess of LREE and notable negative Eu anomaly. Enrichment in Sr and lack of negative Eu anomaly observed for most of the samples (Fig. 7b right), coupled with elevated concentration of Ca (Appendix 1) is explained by the high content of clinopyroxene and/or plagioclase. Exceptions are again Sijarina amphibolites (SM541-1) and Gornji Brestovac meta-gabbro (SM560-1) showing slightly lower Sr concentrations. The negative anomalies of Nb and Ta (Fig. 7b right), and its position on the discrimination diagram in Fig. 6b, suggest a continental-arc environment for the Slatina metagabbro (SM549-1).

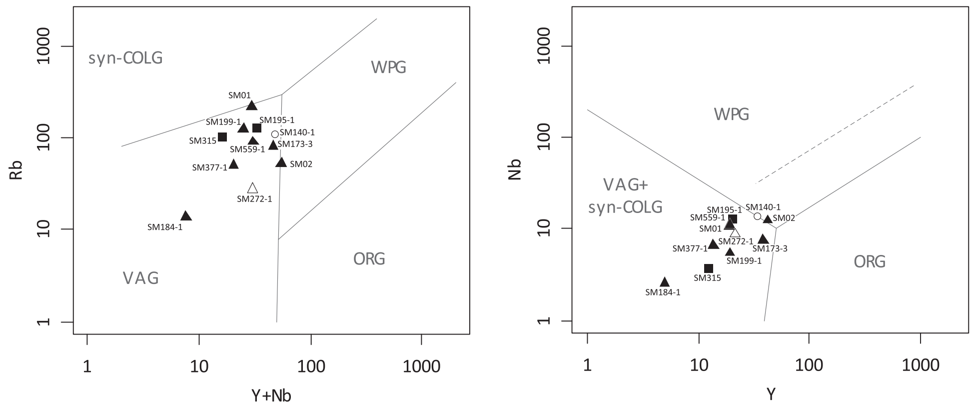


Fig. 4. Tectonomagmatic discrimination diagrams for granites (Pearce et al., 1984). Symbols as in Fig. 3. Abbreviations: Syn-COLG — syn-collisional granites, WPG — within-plate granites, ORG — ocean ridge granites, VAG — volcanic-arc granites.

4.1.2. Vlasina Unit

Acidic and intermediate rocks from Vlasina Unit include a granite (SM272-1), and a diorite (SM493-1) from the Božica magmatic complex, and an albitic gneiss (SM505-1) chemically equivalent to granodiorite (Fig. 3c). All samples from this group are associated with calc-alkaline series (Fig. 3d). Although granite from the Božica complex (SM272-1) shows magmatic-arc signature on the classification diagrams of Pearce et al. (1984) (Fig. 4), it plots in the field of within-plate granites on the discrimination diagram of Harris et al. (1986) (Fig. 6a), due to higher Ta content.

Granites and gneisses from the Vlasina Unit exhibit elevated LREE concentrations with relatively flat HREE pattern and a negative Eu anomaly typical for continental crustal rocks and volcanic-arc granites (VAG) with lack or negligible garnet and zircon fractionation. The Eu anomaly is less pronounced and HREE content is slightly higher in the case of diorite from the Božica complex (SM493-1, Fig. 7c left). It also lacks the pronounced negative Sr anomaly observed in other samples of this group (Fig. 7c right). Both samples from Božica magmatic complex display somewhat lower Rb and K content with elevated Ba concentrations. Similar signature is previously reported for monzogranites and granites in this complex (Vasković and Tasić, 1997).

The basic rocks from the Vlasina Unit are represented by the Lisina gabbro (SM352), deformed monzo-gabro (SM178-1) and a diabase (SM270-3) chemically equivalent to gabbro-diorite (Fig. 3c). The latter two are representing calc-alkaline differentiation series, while the Lisina gabbro (SM352) plots within the tholeiitic series field (Fig. 3d). Based on the classification diagram of Wood (1980), the protoliths of samples SM178-1 and SM270-3 originated from MORB (Fig. 6b), whereas Lisina gabbro (SM352) plots in the field representing within-plate setting.

Similarly to the basic samples of the Lower Complex, basic rocks of the Vlasina Unit also show almost flat REE pattern, with slight depletion in HREE (Fig. 7d left). However, more prominent depletion of HREE is observed for Lisina gabbro (SM352). Content of incompatible alkalis and alkaline earth metals varies strongly, ranging from enriched in SM352 to relatively depleted in diabases SM270-3 (Fig. 7d right).

A single sample of serpentinite (SM502-1) was taken from the Vlasina Unit north of Surdulica granodiorite. It shows high Mg and very low Si content (Appendix 1) and its protolith is equivalent to komatiite according to the geochemical classification of Jensen (1976; Appendix 6). This sample displays depleted and disturbed LREE pattern with pronounced negative Eu anomaly and slightly enriched HREE contents (Fig. 7d left). Contaminated source of the protolith is suggested from the elevated contents of Ba, Ta, Hf and Zr.

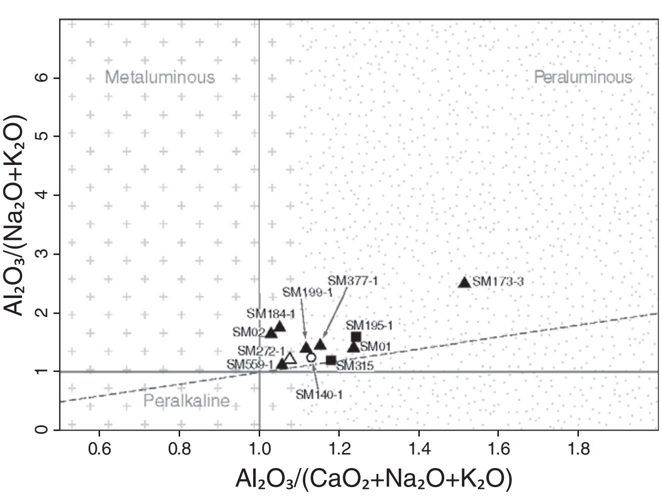


Fig. 5. A/CNK vs. A/NK classification plot (Shand, 1943) for granitic rocks; area stippled with dots is related to S-type granites; area stippled with plus symbols denominates I-type granites. Symbols as in Fig. 3.

4.1.3. Eastern Veles series

The metagranite from the Bukovac locality southwest of Bujanovac (SM315) and granite from the Štip magmatic complex (SM195-1) from the Eastern Veles series belong to the calc-alkaline series (Fig. 3e and f). Although the respective content of Y, Nb and Rb designate these rocks as magmatic-arc granites (Fig. 4), their peraluminous signatures suggest significant crustal input (Fig. 5). Furthermore, based on the Hf–Rb/30–3·Ta systematics of Harris et al. (1986), granite from the Štip complex shows post-collisional signature (Fig. 6a).

Both samples show enrichment in LREE, pronounced negative Eu anomalies and relatively flat HREE patterns, with an exception of a Tm spike for sample SM315 (Fig. 7e left). Accordingly, similar pattern typical for volcanic-arc granites can be observed on P-MORB normalised diagram, with more pronounced negative anomalies in Nb, Ta, Sr and Eu in case of SM315 (Fig. 7e right).

4.1.4. Basement of the Struma Unit

The analysed Delčevo granite (SM140-1) belongs to the Struma Diorite Complex (SDC), while the Bosilegrad monzonite (SM236-1), and a single basic sample (SM231-1) consistent with gabbroic rocks (Fig. 3e), were taken from the COL complex. All of the COL complex samples are associated with calc-alkaline differentiation series (Fig. 3f). Delčevo granite (SM140-1) shows signature typical for magmatic-arc granites (Fig. 6a), however a high affinity to within-plate setting is also suggested by its position on the Nb/Y diagram and by its peraluminous signature (Figs. 4 and 5, respectively). On a geotectonic discrimination plot for basic rocks, COL complex gabbro (SM231-1)

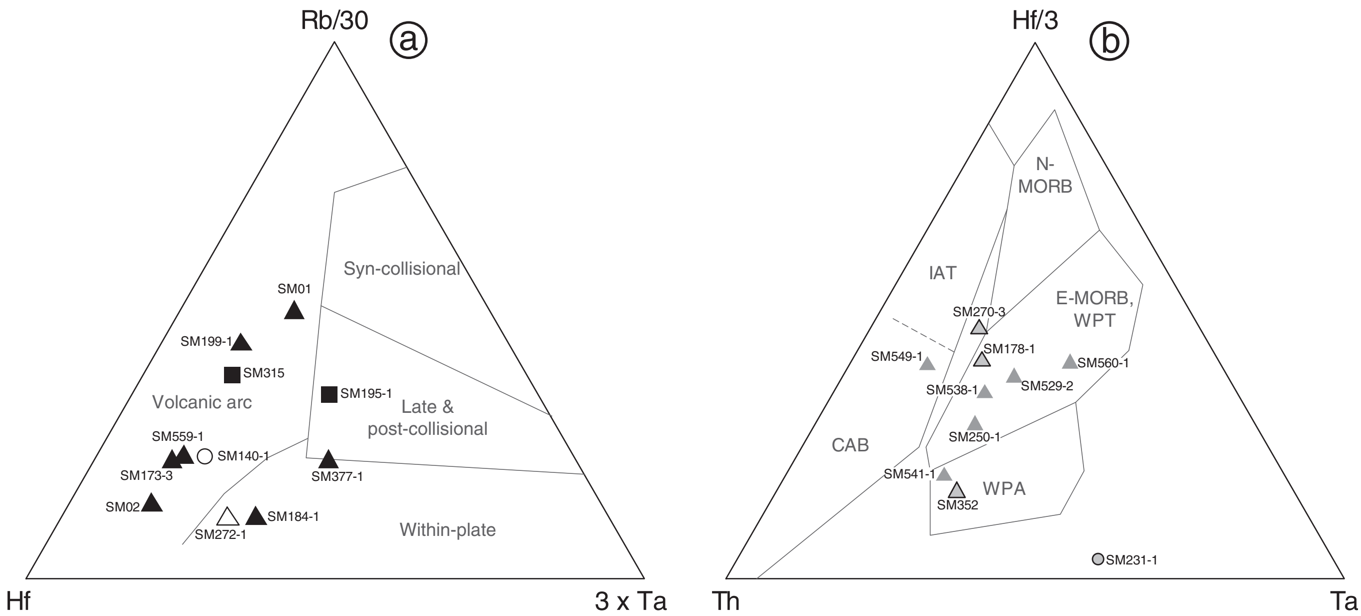


Fig. 6. (a) Ternary geotectonic discrimination diagram for granites (Harris et al., 1986); (b) Tectonomagmatic classification diagram for basic rocks (Wood et al., 1979). Symbols as in Fig. 3. Abbreviations: CAB — continental-arc basalts, IAT — island-arc tholeiits, N-MORB — normal mid-ocean ridge basalts, E-MORB — enriched mid-ocean ridge basalts, WPT — within-plate tholeiits, WPA — within-plate alkaline magma.

plots well outside the within-plate area due to its anomalously high Ta and Hf content (Fig. 6b).

Sample SM140-1 shows an REE pattern with elevated content of LREE followed by slightly negative Eu anomaly and a relatively flat HREE pattern except for the slight enrichment of Yb and Tm (Fig. 7e left). Bosilegrad monzonite (SM236-1) is generally more depleted than the other two samples and displays a positive Eu anomaly and an enrichment in heaviest REE. All three samples exhibit prominent enrichment in Rb, Ba, K, Hf and Zr and a depletion of Ta and Nb, owing to their magmatic-arc origin (Fig. 7e right). Bosilegrad monzonite (SM236-1) displays an elevated Sr content, in which Delčevo granite (SM140-1) is considerably depleted most probably due to lower plagioclase content. Gabbroic sample (SM231-1) shows overall depleted REE pattern, with somewhat higher content of LREE, prominent positive Eu anomaly and gently sloping HREE sequence (Fig. 7d left). Additionally, Rb, Ba, Ta and Zr contents are quite elevated with significant loss in Nb and Hf (Fig. 7d right). Elevated concentrations of Sr and Eu could be explained by increased contents of clinopyroxene and plagioclase.

4.2. Geochronology

In this section only the emplacement ages of the magmatic, and maximum ages of deposition of the para-metamorphic rocks will be presented. For extensive sample descriptions and discussion of measurements see Appendix 3. Interpretation of the detrital and inherited zircon ages is given in Section 5.5.

4.2.1. The Lower Complex

Ten samples of magmatic rocks and two para-metamorphic rocks were taken for age determination in the Lower Complex (Table 1 and Fig. 8). The oldest magmatic body in the Lower Complex is the Vlajna granitoid with an age of 558 ± 6 Ma (SM02, Section 1.1. in Appendix 3). Forty age measurements of detrital zircons from the para-metamorphic rocks in Golemo Selo (SM250-2, Section 1.2.2. in Appendix 3) and Sijarinska Banja (SM310, Section 1.4.2. in Appendix 3) constrain the maximum depositional age close to the emplacement age of Vlajna granitoid (569 ± 9 Ma and 562 ± 6 Ma, respectively). Reactivation of the magmatic activity in the Lower Complex occurred at 490 ± 7 Ma with the emplacement of the leucocratic dykes in Vinica area (SM173-3, Fig. 8, Section 1.3.2. in Appendix 3), and a group of small leucocratic bodies in Vrvi Kobila area at 478 ± 3 Ma (i.e. Kukavica granites; SM01; Fig. 8, Section 1.6.1. in Appendix 3). Orthogneiss in the Maleševski Mts. of the Ograzhden block intruded at 472 ± 4 Ma (SM184-1, Fig. 8, Section 1.4.1. in Appendix 3). Emplacement ages of amphibolites in Golemo Selo (SM250-1, Section 1.2.1. in Appendix 3), and Vinica area (SM173-1, Section 1.3.1. in Appendix 3), were determined at 462 ± 6 Ma and 456 ± 2 Ma, respectively. Zircons from the deformed coarse-grained Qz monzonite in the southwestern periphery of the Bujanovac pluton gave an emplacement age of 439 ± 2 Ma (SM377-2, Section 1.5.3. in Appendix 3). Late Variscan magmatic activity in the Lower Complex is represented by rather small, undeformed Slatinska Reka granite intruded at 328 ± 5 Ma (SM550-1, Section 1.6.2. in Appendix 3). Two samples from the fine-grained Bujanovac pluton represent the youngest magmatic activity revealed in the Lower Complex with emplacement age of 253 ± 2 Ma in the central part (SM199-1, Section 1.5.1. in Appendix 3), and 255 ± 3 Ma in the southwestern periphery of the pluton (SM377-1, Section 1.5.1.2. in Appendix 3).

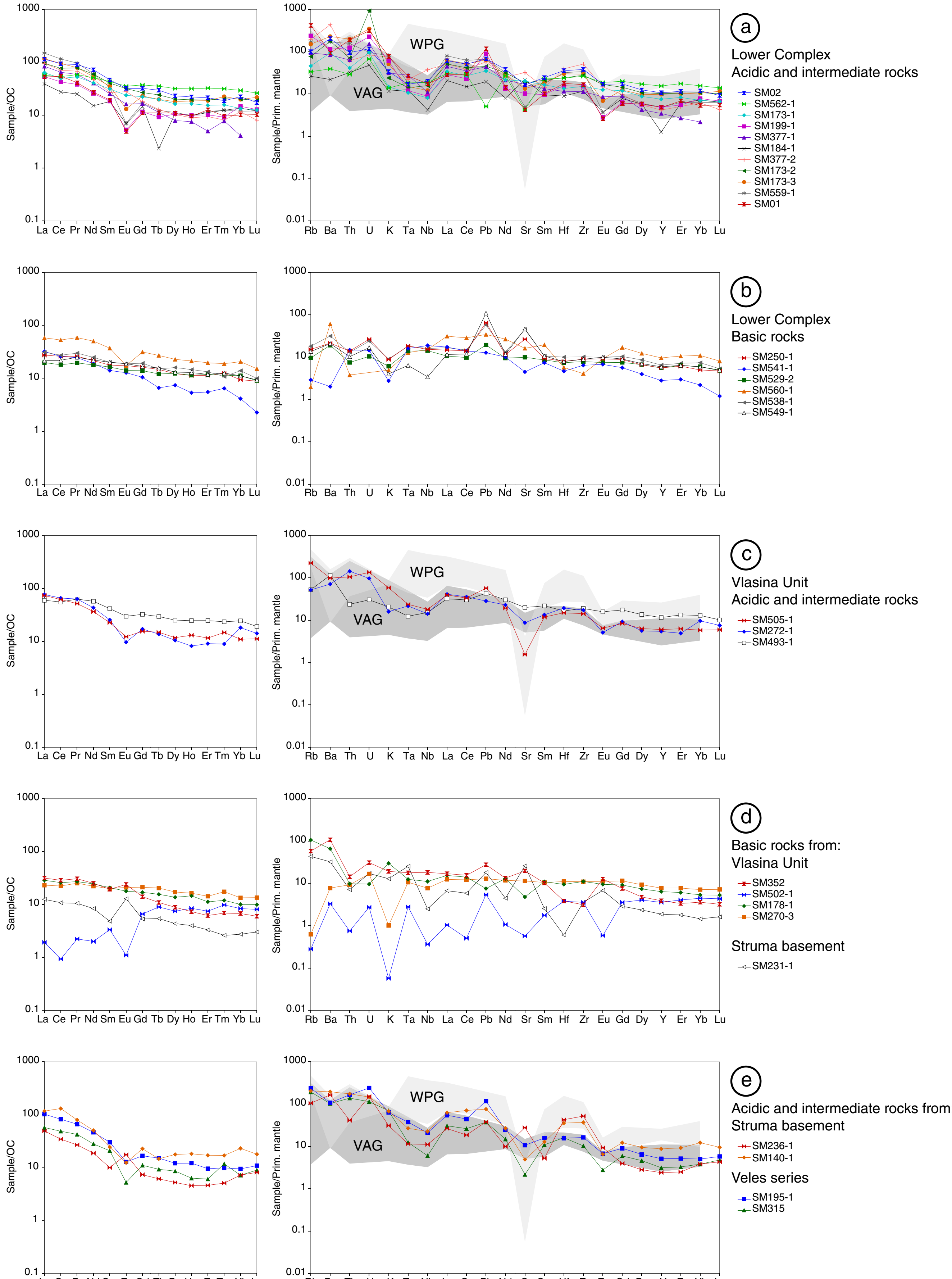
Most of the single grain U–Pb ages in the Lower Complex are Late Ordovician, but the duration of magmatic activity lasted from late Cambrian to earliest Silurian (Fig. 9). Late Neoproterozoic, late early Carboniferous and Permian–Triassic zircon ages are also present.

4.2.2. Vlasina Unit

Only three samples from Vlasina Unit yielded zircon grains of sufficient size and quantity for LA-ICP-MS U–Pb measurements. The age results derived from these measurements show that magmatic activity in Vlasina Unit lasted from late Neoproterozoic until early Cambrian (Fig. 9). The Doganica metagranite is the oldest magmatic body in the Vlasina Unit with an emplacement age of 562 ± 2 Ma (SM600-1, Fig. 8, Section 2.1. in Appendix 3). The slightly younger Lisina gabbro was emplaced at 550 ± 11 Ma (SM3352, Section 2.3. in Appendix 3). Finally, a granitoid within the Božica magmatic complex was emplaced at 521 ± 4 Ma (SM272-1, Section 2.2. in Appendix 3).

4.2.3. Eastern Veles series

Crystallisation ages of zircon domains from the rocks of the Eastern Veles series resemble those obtained from the Lower Complex (Fig. 9).



Sample/OC
Sample/Prim. mantle
1000
100
10
1
0.1
0.01
La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
Rb Ba Th U K Ta Nb La Ce Pb Nd Sr Sm Hf Zr Eu Gd Dy Y Er Yb Lu
WPG
VAG
a
Lower Complex
Acidic and intermediate rocks
SM02
SM562-1
SM173-1
SM199-1
SM377-1
SM184-1
SM377-2
SM173-2
SM173-3
SM559-1
SM01
b
Lower Complex
Basic rocks
SM250-1
SM541-1
SM529-2
SM560-1
SM538-1
SM549-1
c
Vlasina Unit
Acidic and intermediate rocks
SM505-1
SM272-1
SM493-1
d
Basic rocks from:
Vlasina Unit
SM352
SM502-1
SM178-1
SM270-3
Struma basement
SM231-1
e
Acidic and intermediate rocks from:
Struma basement
SM236-1
SM140-1
Veles series
SM195-1
SM315

The earliest magmatic activity is represented by the Bukovac leuco-granite with an emplacement age of 487 ± 17 Ma (SM315, Fig. 8, Section 3.1. in Appendix 3). A sample of biotite-rich granites from the Štip magmatic complex has a weighted average of the youngest group of age-results of 304 ± 3 Ma (SM195-1, Fig. 8, Section 3.3. in Appendix 3). Seventeen age-measurements of detrital zircons from the Novo Brdo schists allowed only tentative constraints on the maximum deposition age of its sedimentary protolith at 255 ± 2 Ma (KOS02, Fig. 8, Section 3.2. in Appendix 3).

4.2.4. Basement of the Struma Unit

Similarly to Vlasina Unit, the magmatic record in the COL complex and SDC in the basement of the Struma Unit reveal igneous activity lasting from the late Neoproterozoic to the early Cambrian (Fig. 9). The granites from the Delčevo magmatic complex (part of SDC) were emplaced at 536 ± 7 Ma (SM140-1, Fig. 8, Section 4.2. in Appendix 3). A slightly younger age of 522 ± 4 Ma was determined for the Bosilegrad monzonite in the COL complex (SM236-1, Fig. 8, Section 4.1. in Appendix 3).

4.3. Lu–Hf isotopic composition

Deviation of the Hf isotopic composition of zircon grains from the chondritic uniform reservoir (CHUR) reported in age-corrected epsilon units is a widely used parameter for description of the magma sources from which the zircons have crystallised, thus providing an estimate of the significance of juvenile depleted mantle (DM) or reworked old crust during the formation of the melt (e.g. Amelin et al., 1999; Kinny and Maas, 2003). In our study a total of 112 zircon spots were measured for their Lu–Hf isotopic composition (Appendix 7 and Fig. 10a). Details concerning the calculation of εHf values and DM model ages are provided in Appendix 7.

The vast majority of the analysed spots (101 of total 112) are related to a group of zircons that crystallised from the Neoproterozoic to the earliest Triassic (Fig. 10a). Four groups of results younger than 1 Ga are defined based on U–Pb ages of the analysed zircon domains (Fig. 10b). Group D comprises measurements made on Ediacaran to early Cambrian zircon domains, which show a range of εHf values between +12.6 and −2.8 (Fig. 10b). Zircon grains from Vlajna granite (SM02), Bosilegrad monzonite (SM236-1), and granites from Božica (SM272-1), and Delčevo (SM140-1) magmatic complexes show more positive values (+12.6 to +4.2) indicating predominantly juvenile magma source, while the εHf values between +3.7 and −2.8 of zircons from Doganica granite (SM600-1) and Lisina gabbro (SM352) suggest higher presence of crustal material. Zircons with ages corresponding to this age-group are found as detrital grains in Novo Brdo schists (KOS02) and as xenocrysts in Slatinska Reka granite (SM550-1), Kukavica granite (SM01), and amphibolites (SM173-1) and leucocratic dykes in Vinica area (SM173-3). Group C contains measurements made on zircons that crystallised between the late Cambrian and the early Silurian (Fig. 10b). The εHf values of these zircon domains range between +18.7 and −6.2. Zircons from Kukavica granite (SM01), coarse-grained Bujanovac Qz-monzonite (SM377-2), Maleševski Mts. orthogneiss (SM184-1) represent part of the group C with lower εHf values (+4.3 to −6.2) indicating mixed juvenile and continental crust signature of the melt. Zircons of similar age and range of εHf values occur as detrital grains in Novo Brdo schists (KOS02) and as xenocrysts in fine-grained Bujanovac granite (SM199-1), granite from the Štip complex (SM195-1) and amphibolites from Vinica area (SM173-1). Formation of new crust associated with age-group C is documented by zircons with εHf values between +18.7 and +6.9, from amphibolites and leucocratic dykes in Vinica area (SM173-1 and SM173-3, respectively), and Bukovac leuco-granite (SM315). Xenocrysts from the Golemo Selo amphibolite (SM250-1) and Maleševski Mts. orthogneiss (SM184-1) yielded similarly high εHf values. Group B is composed of zircon domains crystallised in Devonian and Carboniferous (Fig. 10b). Low to negative εHf values of these zircons (+2.6 to −11.2) suggest crystallisation in magma predominantly related to reworking of older continental crust. Group B includes zircons of similar age and εHf values from the granite in the Štip magmatic complex (SM195-1), Slatinska Reka granites (SM550-1), and zircon domains from the Novo Brdo schists (KOS02). Zircons from fine-grained Bujanovac granite (SM199-1 and SM377-1) constitute the youngest group A (Fig. 10b), with crystallisation ages ranging from the late Permian to the Early Triassic. High εHf values of these zircon domains +19.7 and +8.5 suggests crystallisation from a juvenile magma source. Permian–Triassic age of the youngest detrital grain of Novo Brdo schists (KOS02) fits within the group A. However, its εHf value of −7.3 is in contrast to very high values of zircons from the Bujanovac granite, indicating a source area exotic to the SMM. The presence of late Neoproterozoic inherited zircon cores, detrital grains and xenocrysts with εHf values between +7.5 and −18.3 (580–694 Ma; Fig. 10b), suggests that the hypothetical basement of the magmatic and sedimentary rocks of the SMM, originated as a mixture of reworked crust and DM-derived magmas, comparable to the Neoproterozoic arc at the northern margin of Gondwana (e.g. Linnemann et al., 2007).

The negative εHf value (−3.3) and the resulting DM model age (T_{DM}) of 3.4 Ga of the single oldest inherited core (Fig. 10a and Appendix 7; SM250-2-o19; 3049 Ma) suggest that this initially detrital grain could be derived from a reworked Archean crust. Range of εHf values of Siderian–Rhyacian zircon domains (2.4–2.2 Ga; Fig. 10a) reveal that the early Palaeoproterozoic crust comprised both reworked Archean crust and DM sources. The DM model ages of the two spots with the lowest epsilon values correspond to the crustal evolution trend for the Siderian–Rhyacian juvenile crust (ca. 2.2 Ga). However these T_{DM} could also result from mixing of minor quantities of juvenile material at 580 Ma with large volumes of partial melts of an Archean crust. Four late Neoproterozoic to late Mesoproterozoic (from 586 to 1075 Ma; Appendix 7 and Fig. 10a) cores from magmatic zircons with negative epsilon values between −3.56 and −12, are aligned along a line subparallel to the crustal evolution trend (Fig. 10a). Their DM model ages (between 1.81 and 1.9 Ga) would suggest existence of an Orosirian juvenile crust, although they could also be related to reworking of an older crustal segment at, or prior to 1075 Ma. A group of 17 Cryogenian to Triassic zircons (694–255 Ma) with negative epsilon values and a single εHf-positive Stenian grain (1177 Ma) are roughly aligned along a linear trend with T_{DM} between 1.3 and 1.5 Ga (Fig. 10a and Appendix 7). Although similar DM model ages were determined for the Neoproterozoic granites of the Menderes Massif (Zlatkin et al., 2013), their parent-magma is suggested to represent a mixture of sources with different isotopic compositions.

5. Discussion

The results of our research suggest the existence of four major stages of magmatic activity within the Serbo-Macedonian Massif and the adjacent basement units (Figs. 8 and 9): i) Ediacaran to mid-Cambrian (late Cadomian; 562–521 Ma), ii) late Cambrian to early Silurian (490–439 Ma), ii) Carboniferous (late Variscan; 328–304 Ma), and iii) late Permian to Early Triassic (Permian–Triassic; 255–253 Ma).

Fig. 7. Trace element pattern plots normalised for chondritic (Nakamura, 1974) and primitive mantle concentrations (Hofmann, 1988). Shaded areas WPG and VAG represent envelope concentrations of within-plate and volcanic-arc granites, respectively (Pearce et al., 1984). OC stands for ordinary chondrite. In (e) and (d) basement of Struma Unit includes samples from both the COL complex and the SDC.

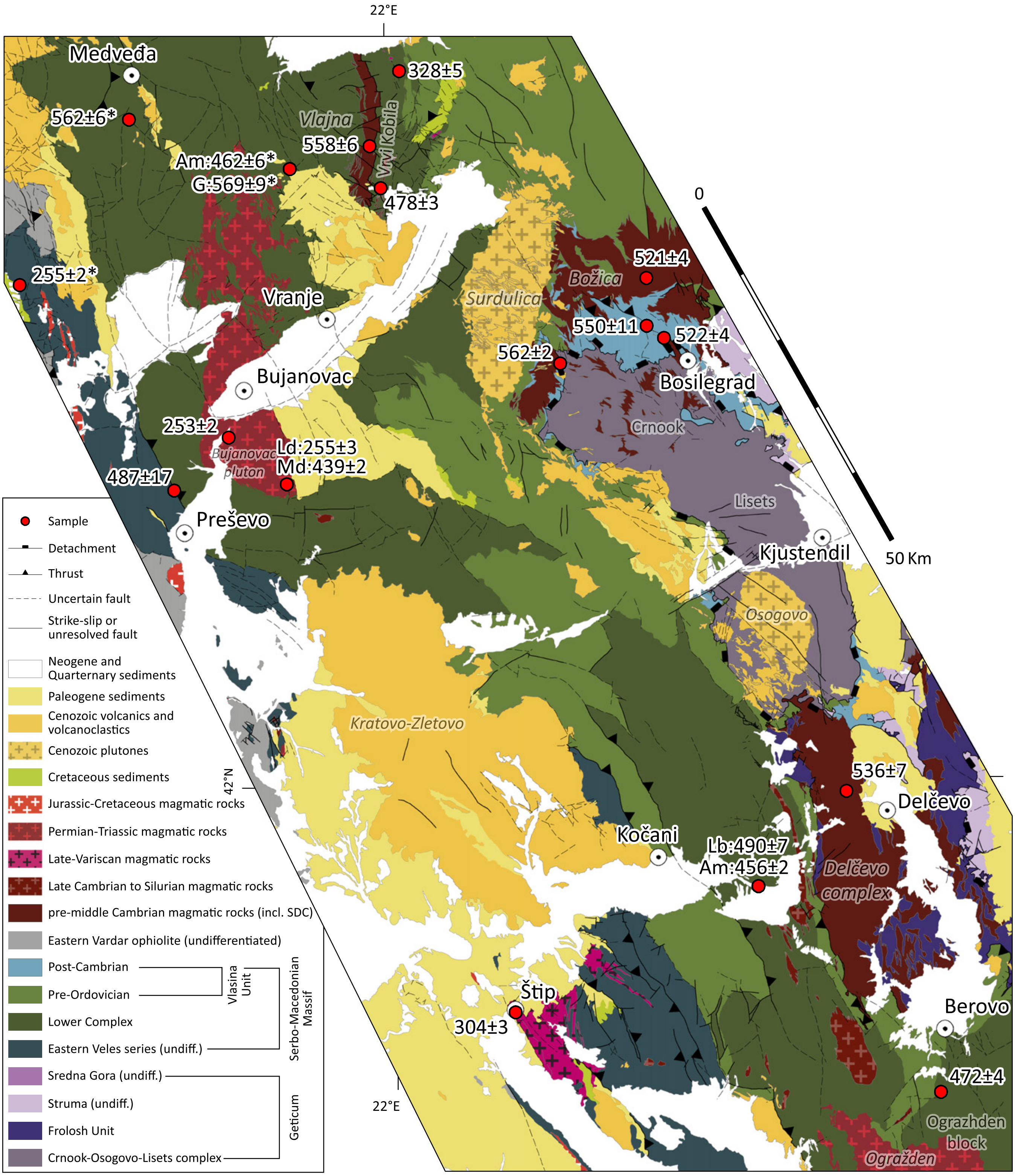


Fig. 8. Tectonic map of the study area with the $^{206}Pb/^{238}U$ age results including 2σ errors. Ages reported with an asterisk (*) were determined based on detrital or xenocrystic zircon content. Abbreviations: Am — amphibolite, G — gneiss, Lb — leucocratic dykes, Md — more deformed, Ld — less deformed.

5.1. Late Cadomian magmatic stage (Ediacaran to mid-Cambrian, 562–521 Ma)

Magmatic bodies emplaced during the Ediacaran to mid-Cambrian stage are present in all studied tectonic units (phase D in Fig. 9). This phase could be separated in two substages (562–550 Ma and 536–521 Ma) separated by a short period of quiescence. This magmatic phase could correspond to the Cadomian orogeny which is generally manifested by a series of tectonic events which took place along the northern margin of Gondwana from ca. 750 to 540 Ma (Nance et al., 2002; Murphy et al., 2004; Linnemann et al., 2007).

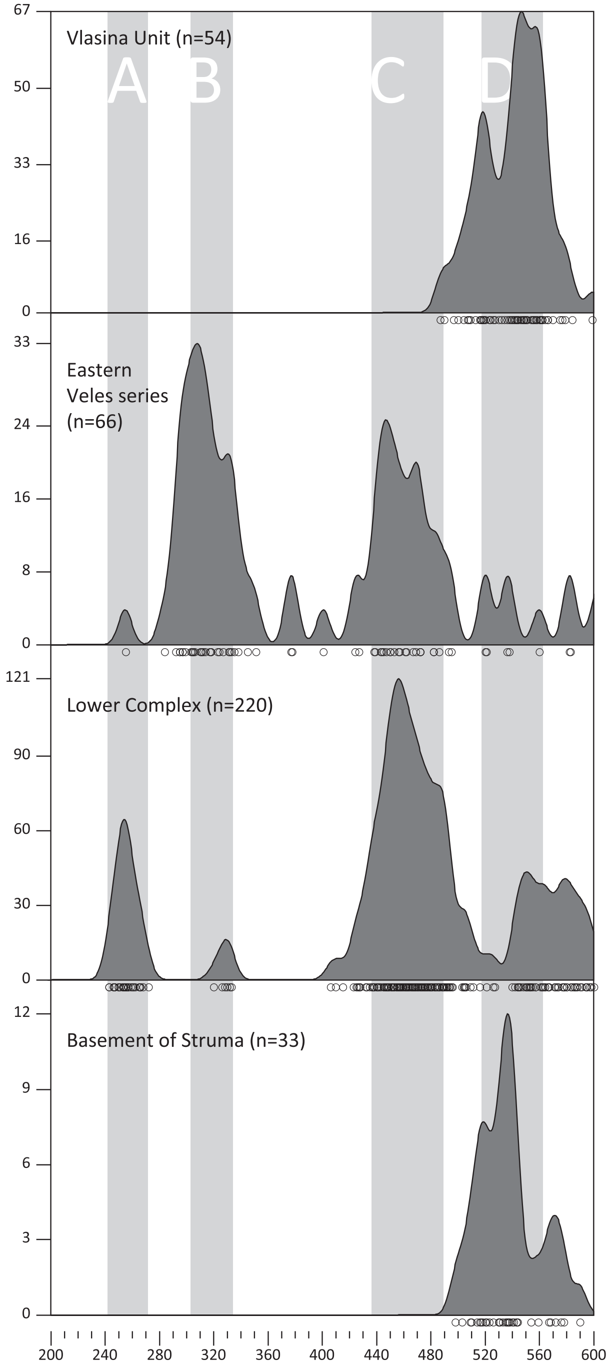


Fig. 9. Kernel density estimates of $^{206}Pb/^{238}U$ age results between 200 and 600 Ma, related to tectonic units. Age results considered include both the measurements from detrital and magmatic zircons. Letters A, B, C, and D correspond to different magmatic stages.

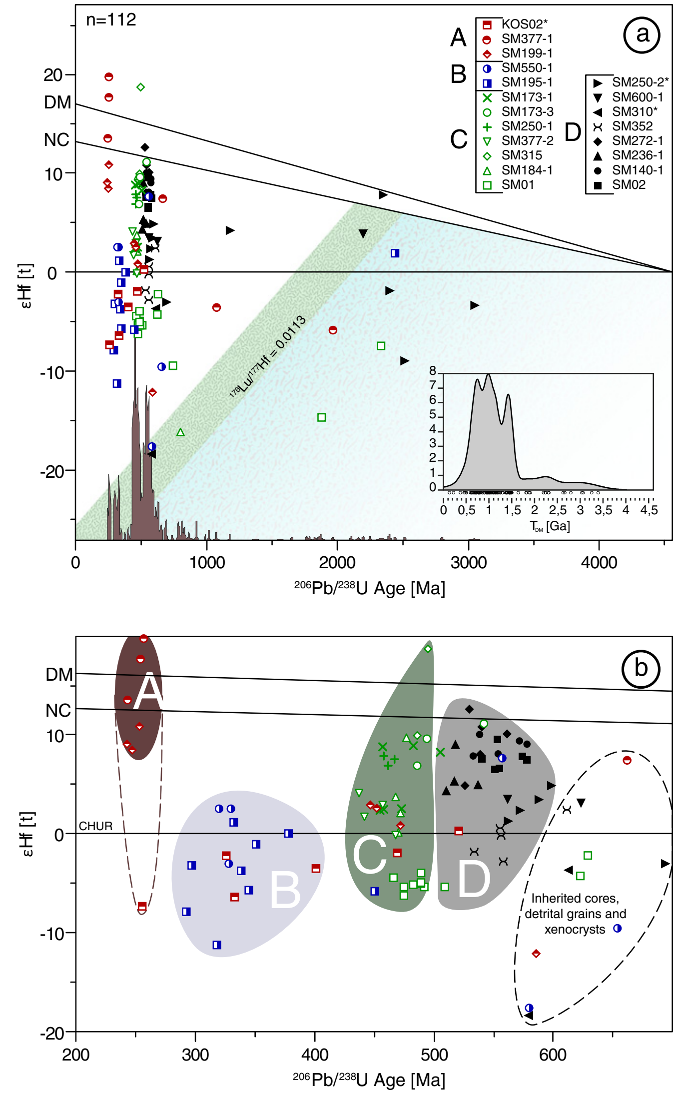


Fig. 10. (a) Hf isotope evolution diagram of all analysed zircons. Samples with detrital zircons are marked with an asterisk. Evolution curves for the new crust (NC) and depleted mantle (DM) after Dhuime et al. (2011). CHUR stands for chondritic uniform reservoir. Crustal evolution trend ($^{176}Lu/^{177}Hf = 0.0113$) after Linnemann et al. (2014). Stippled fields represent Siderian–Rhyacian and Archean crustal trends (from left to right, respectively). Probability density plot at the bottom of the diagram was produced by AgeDisplay (Sircombe, 2004), and represents all age results. Inset contains Kernel density estimate of DM model ages produced by DensityPlotter (v 6.1; Vermeesch, 2012). See text and Appendix 7 for details. (b) Magnified area between 200 and 700 Ma from (a).

The older episode is represented by the emplacement of the Vlajna granite (SM02; 558 ± 6 Ma) in the Lower Complex, and Doganica metagranites (SM600-1; 562 ± 2 Ma) and Lisina gabbro (SM352; 550 ± 11 Ma) in the Vlasina Unit. Geochemical signatures of Vlajna granite and Lisina gabbro indicate formation in convergent setting and contamination with crustal material (Figs. 4, 6b, and 7a and d).

The continuation of the magmatic activity in the early Cambrian includes intrusion of peraluminous Delčevo granite (SM140-1; 536 ± 7 Ma) of the Struma Diorite Complex into the ophiolites of Frolosh Unit, the Božica complex (SM272-1; 521 ± 4 Ma) into the Vlasina Unit, and the Bosilegrad monzonite (SM236-1; 522 ± 4 Ma) into the COL complex. The geochemical signatures of samples from the Božica magmatic complex and the magmatic rocks of the COL complex are consistent with mainly magmatic-arc setting (Figs. 4, 6a, and 7c and e).

Magmatic rocks of similar age have been reported in the basement of Struma Unit in Bulgaria (Zagorchev et al., 2011a; Kounov et al., 2012), in the magmatic basement of the Pirgadikia Unit in the Internal Hellenides (Himmerkus et al., 2007, 2009a), and in the Neoproterozoic basement of the Sebeş–Lotru Unit in the southeastern Carpathian mountains (Balintoni et al., 2010a; Balintoni and Balica, 2013a). Additionally, coeval Ediacaran to early Cambrian I-type arc-related magmatism was determined in the İstanbul (Chen et al., 2002; Ustaömer et al., 2005) and Istranca (Yilmaz Şahin et al., 2014) terranes of the Western Pontides.

5.2. Late Cambrian to early Silurian magmatic stage (490–439 Ma)

An apparent 30 Ma of quiescence are followed by a long period of recurrent magmatic activity throughout the Lower Complex and the

Eastern Veles series dominated by peraluminous granites and mafic intrusions (phase C in Fig. 9). Three pulses could be distinguished within this stage. The oldest pulse is represented by leucocratic dykes from the Vinica area of the Lower Complex (SM173-3; 490 ± 7 Ma), Bukovac leucocratic metagranite from the eastern margin of the Eastern Veles series (SM315; 487 ± 17 Ma), and Kukavica granites (SM01; 478 ± 3). All these igneous rocks are peraluminous and have prevalent magmatic-arc signature (Figs. 4, 5, 6a, and 7a and e), with the Kukavica granites showing substantial enrichment in crustal material (Fig. 10). Similar age of emplacement was obtained for the Lower Complex orthogneiss in Maleševski Mts. at the southeastern margin of the study area (SM184-1; 472 ± 4 Ma). Although they share a dominant magmatic-arc geochemical signature, this orthogneiss exhibits a clear I-type signature and within-plate affinity (Figs. 5 and 6a). Meta-granites from the Bulgarian part of the same mountain range yielded Late Ordovician to early Silurian ages (476–433 Ma) and nearly identical geochemical signature (Zidarov et al., 2003a,b; Macheva et al., 2006; Zagorchev et al., 2011a; Kounov et al., 2012). The Bretila and Cumpăna units of the Carpathian Mts. in Romania comprise arc-related magmatic rocks within the same age range (Balintoni et al., 2010a; Balintoni and Balica, 2013b). Slightly younger age (443–426 Ma) and magmatic-arc origin was determined for the peraluminous granites and orthogneisses of the Vertiskos unit in Greece (Himmerkus et al., 2007, 2009a; Meinhold et al., 2010).

The second magmatic pulse is marked by mafic intrusions in the Lower Complex. Zircon xenocrysts from an amphibolite near Golemo Selo in the Lower Complex yielded Middle Ordovician ages (SM250-1; ca. 462 Ma). These amphibolites represent dykes of within-plate tholeiitic basalts most probably intruded during an early episode of continental rifting, and later deformed together with the sedimentary host rocks. Compatibility in trace-element concentrations with undated amphibolites and meta-gabbros elsewhere along the Lower Complex (Fig. 7), together with rocks of similar chemistry north of the study area (Cvetković, 1992; Karamata and Krstić, 1996), suggest a regional importance of this rifting event. Nearly coeval age of emplacement was determined for amphibolites in the Vinica area of the Lower Complex (SM173-1; 456 ± 2 Ma), however their trace element content points to arc-related setting. Higher concentrations of large-ion lithophile elements (LILE) and LREE concentrations (Fig. 7) and wider range of (positive) εHf values (Fig. 10b) suggest higher amount of crustal material involved in the formation of the Vinica amphibolite compared to the other analysed mafic bodies from the Lower Complex.

The final magmatic pulse during the stage C (Fig. 9) took place in the earliest Silurian when the medium to coarse-grained Bujanovac Qz-monzonite was intruded in the Lower Complex (SM377-2; 439 ± 2 Ma). High content of elongated zircons (Pupin, 1980), low positive εHf values (0 to +4.3, Appendix 7and Fig. 10b), and slightly elevated Eu content (Fig. 7a), suggest that these rocks were formed by primary mantle-derived magma that was significantly contaminated by crustal material or by partial melting of the arc-related crust.

5.3. Late Variscan magmatic phase (Carboniferous; 328–304 Ma)

Although only two magmatic bodies yielded Carboniferous ages in the study area, their emplacement diverges both temporally and in terms of geochemical signature (phase B in Fig. 9). The undeformed Slatinska Reka granite (SM550-1; 328 ± 5 Ma) intruded the gneisses of the Lower Complex, providing the upper age-limit of the high-strain event(s) that affected the host rocks (Fig. 11a). Given the early Silurian age of the youngest dated highly deformed igneous rocks in the SMM (deformed Kukavica granite, SM01; 478 ± 3 Ma), the high strain deformation and peak metamorphism is most probably related to the Variscan orogeny. The nearest magmatic rocks of similar age (334 ± 1 Ma) are the undeformed post-collisional I-type granitoids in the Tran region in Bulgaria (Peytcheva et al., 2009a; Dyulgerov et al., 2010).

The undeformed granite of the Štip magmatic complex in the Eastern Veles series was emplaced during the late Carboniferous (SM195-1;

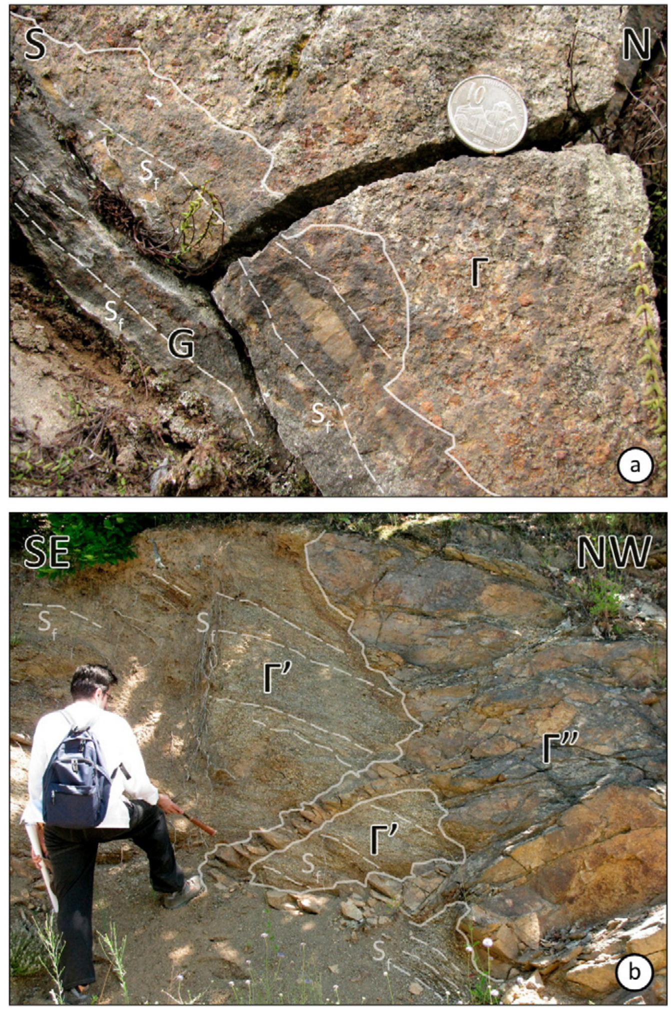


Fig. 11. (a) Slatinska Reka valley (22°1′30.716″E, 42°50′53.365″N). Contact of the Lower Complex gneiss (G) with undeformed Slatinska Reka granite (Γ). Dashed white line indicates the trace of foliation (S_f) in the gneiss. Coin diameter 26 mm; (b) Spančevac locality (21°50′2.569″E, 42°21′33.025″N). Weakly deformed Bujanovac granite (Γ″) intruding the penetratively deformed coarse-grained Qz-monzonite (Γ′) of the Lower Complex. Dashed white lines indicate the trace of foliation (S_f) in the coarse grained Qz-monzonite.

304 ± 3 Ma). Previously reported Callovian–Oxforidan Rb–Sr age of biotite (Šoptrajanova, 1967) is most probably related to thermal overprint (Spray et al., 1984), or to a much younger magmatic body that was intruded in the Carboniferous pluton. Large number of inherited zircons, peraluminous character of the granite (Fig. 5), and a wide range of low εHf values (−11.2 to +2, Fig. 10) suggest that the magmatic-arc geochemical signature (Fig. 4) is probably an artefact of the arc-related volcano-sedimentary basement, that was partially melted during the generation of the source magma of the Carboniferous granite in the Štip magmatic complex. The geochemical affinity to late- or post-collisional setting (Fig. 6a), most probably represents the correct geotectonic setting of this granite. Similar ages were obtained from granitoids in the Balkan terrane (Carrigan et al., 2005), and Danubian and Getic nappes of the Carpatho-Balkanides (Duchesne et al., 2008; Vasković et al., 2012).

5.4. Late Permian to Early Triassic magmatic phase (255–253 Ma)

The fine-grained Bujanovac granite (SM377-1 and SM199-1) was emplaced within the deformed medium- to coarse-grained Silurian Bujanovac Qz-monzonite (SM377-2, Fig. 11b) and the gneisses of the Lower Complex at the end of Permian and beginning of Triassic (SM377-1: 255 ± 3, and SM199-1: 253 ± 2 Ma). Magmatic-arc geochemical signature of the central part of the fine-grained Bujanovac pluton (SM199-1; Figs. 4, 6a and 7a) could be related to the extensive contamination by reworked lower crustal material of arc-related origin, rather than geotectonic setting during the magma generation. This interpretation is supported by an abundance of inherited zircons of Early Palaeozoic and Cadomian age and a peraluminous signature of the Bujanovac granite (Figs. 5 and 12). Within-plate affinity interpreted for the sample from the southern periphery of the pluton (SM377-1) is presumed to be more representative for the Permian-Triassic Bujanovac pluton (Fig. 6a). Furthermore, positive εHf values (+8.5 to +19.7) are associated with the significant influence of the depleted mantle on the source of late Bujanovac granites (Fig. 10a and b). Such contamination with juvenile material could be caused by mafic intrusions in the severely attenuated continental crust during the rifting stage leading to the opening of the Mesozoic Tethys (Karamata and Krstić, 1996; Karamata, 2006).

Intrusions of similar age have been reported in other parts of the SMM, such as rift-related Arnea and Kerkini A-type granites in Vertiskos (Christofides et al., 1999, 2007; Himmerkus et al., 2009b), and Igralishte, Skrat (Skrut) and Ogražden granites in the Ograzhden block (Zidarov et al., 2004; Peytcheva et al., 2005; Zidarov et al., 2007; Peytcheva et al., 2009b; Georgiev et al., 2012).

5.5. Detrital and inheritance record and implications on the provenance of the SMM

Age results of the detrital zircons from the para-metamorphic samples allowed determination of relative depositional age of the sedimentary protoliths. The maximum depositional age of the sedimentary protoliths of the paragneisses within the volcano-sedimentary Lower Complex could be constrained as late-Cadomian from the youngest detrital zircons obtained from the Sijarinska Banja and Golemo Selo paragneiss (SM310; 562 ± 6 and SM250-2; 569 ± 9 Ma, respectively). Although much younger (Meinhold et al., 2010), the sedimentary cover of the Pirgadikia Unit of the internal Hellenides shares much of its Precambrian detrital record with Golemo Selo paragneiss of the SMM (Fig. 12).

The age of the Vlasina volcano-sedimentary successions could not be determined directly since meta-sedimentary rock samples collected for this purpose did not yield sufficient number of zircon grains of appropriate size for an LA-ICP-MS analysis. However, the early Cambrian age of emplacement of a granite in the Božica magmatic complex constrains the youngest age of Vlasina Unit, since the xenoliths found within the Božica complex in Bulgaria are suspected to be derived from the pre-

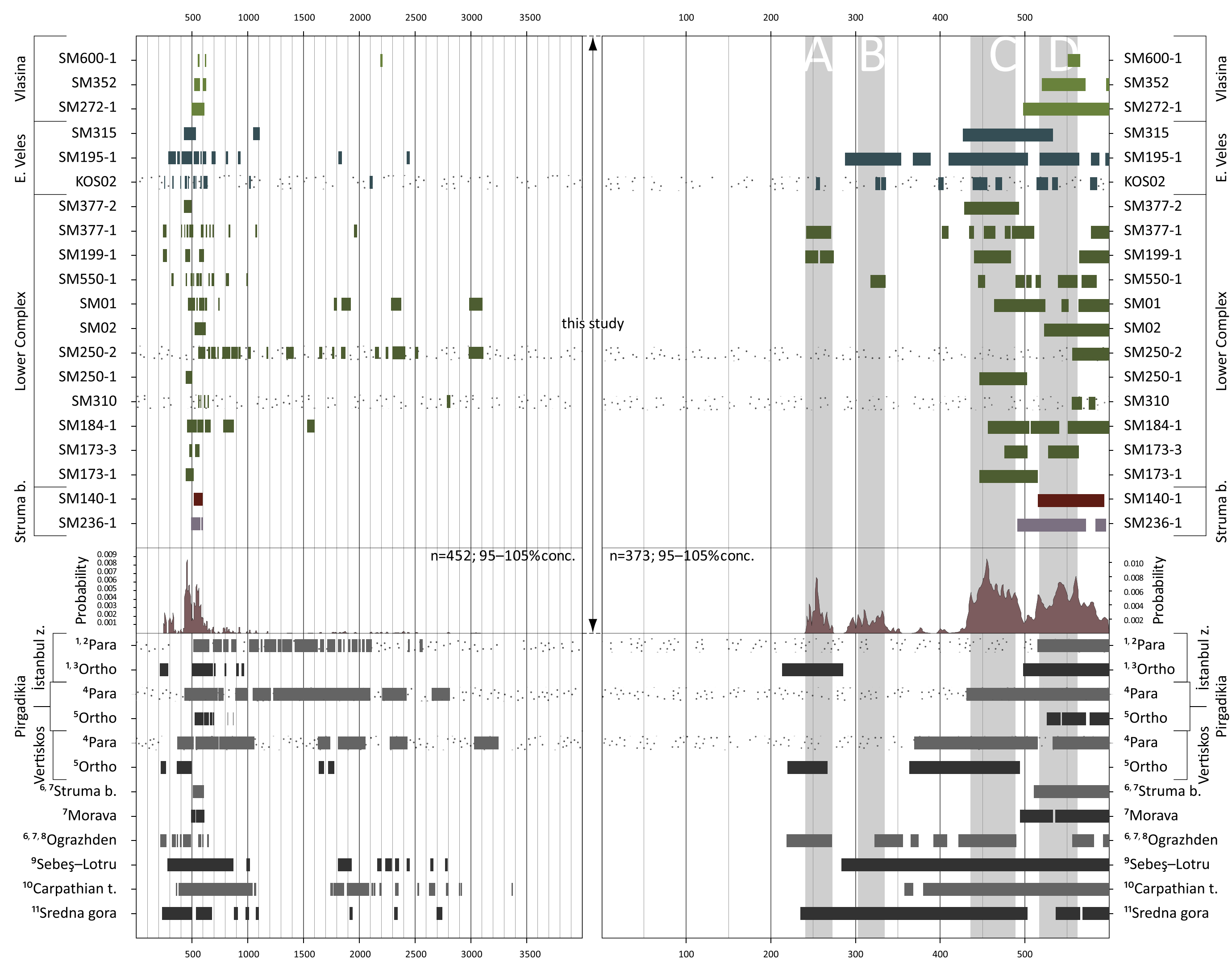


Fig. 12. Zircon-age content of notable occurrences in the pre-Mesozoic basement in southeastern Europe and the samples from this study. These are delimited by synoptic density distribution of results from this study produced by AgeDisplay (Sircombe, 2004). Presented data includes magmatic, metamorphic and detrital zircons including 2σ errors. Right side of the diagram displays data until 600 Ma. Dot-stippled fields denote samples of sedimentary origin. Sources: [1]Chen et al. (2002); [2]Ustaömer et al. (2011); [3]Ustaömer et al. (2005); [4]Meinhold et al. (2010); [5]Includes: Himmerkus et al. (2006), Himmerkus et al. (2009a), Christofides et al. (2007) and Himmerkus et al. (2009b); [6]Zagorchev et al. (2011b); [7]Kounov et al. (2012); [8]Includes: Zidarov et al. (2003a), Zidarov et al. (2003b), Zidarov et al. (2004), Peytcheva et al. (2005), Macheva et al. (2006), Zidarov et al. (2007), Peytcheva et al. (2009b), Haydoutov et al. (2010) and Zagorchev et al. (2011a); [9]Balintoni et al. (2010a); [10]Balintoni and Balica (2013b); [11]Includes: Carrigan et al. (2005) and Carrigan et al. (2006).

Ordovician Vlasina (Graf, 2001). Additionally, the Cadomian ages of Doganica and Lisina magmatic bodies demand a reassessment of the originally proposed intrusive relationship with the Lower Ordovician meta-sediments (Babović et al., 1977; Pavlović, 1977), as they clearly represent inliers within this para-metamorphic succession.

Although based on scarce zircon content (Appendices 3 and 4), the maximum depositional age of the Novo Brdo schists from the sedimentary cover in the northwestern periphery of the Eastern Veles series was tentatively constrained by the youngest result of 255 ± 2 Ma obtained on a zircon fragment with oscillatory zoning (Appendix 3). This detrital age is in agreement with the Triassic age proposed by Pavić et al. (1983), based on the fossil record.

The range of ages of detrital grains, inherited cores and xenocrysts from samples of sedimentary and magmatic origin in the central SMM provides some information on the initial palaeogeographic position of the basement. Late Ediacaran (562–550 Ma) igneous rocks were determined in the Lower Complex, Vlasina Unit and the basement of the Struma Unit, with Cadomian (ca. 700–ca. 540 Ma) magmatic zircons as preponderant inherited population in these units and the Eastern Veles series (Figs. 9 and 12). This prominent population of U–Pb zircon ages coupled with geochemical results, suggests that the basement of the SMM was most probably involved in the Cadomian orogeny as a segment within the active continental margin of north Gondwana (Nance et al., 2002; Neubauer, 2002; Murphy et al., 2004; Stampfli et al., 2013). In this context, the presence of Early Cryogenian to Tonian zircons (677 to 994 Ma, Fig. 12), could be related to the Cadomian part of the arc (Linnemann et al., 2007; Zulauf et al., 2007; Avigad et al., 2012; Pereira et al., 2012a,b; Linnemann et al., 2014), or as detrital input related to the amalgamation of the South-China craton (Cawood et al., 2013; Zhang et al., 2013). The Avalonian provinces are virtually devoid of this age signature since this arc accreted to Gondwanan active margin at ca. 665–650 Ma (Nance et al., 2008). Additional proof excluding Avalonian provenance is the general scarcity of magmatic zircons of Mesoproterozoic age (ca. 1600–1000 Ma). Presence of Mesoproterozoic zircons is commonly related to the orogenic magmatism affecting Laurentia, Baltica or Amazonia (Nance and Murphy, 1996). Thus they are absent from the South China (Zhang and Zheng, 2013) and West African cratons (Nance and Murphy, 1994; Friedl et al., 2000). However, minor population of late Mesoproterozoic zircons determined in samples from the central SMM (Fig. 9 and 12) does not necessarily imply Avalonian provenance, as similar ages were reported for volcanic rocks of the western Yangtze block (South-China craton; Zhang and Zheng, 2013, and references therein), from the Arabian–Nubian Shield and its cover sequence (e.g. Dixon, 1981; Avigad et al., 2003; Be'eri-Shlevin et al., 2012), and from Neoproterozoic and Palaeozoic siliciclastic sediments of southern Libya (Meinhold et al., 2011). Minor presence of this population throughout the Cadomian domain (Linnemann et al., 2007; Pereira et al., 2012a; Linnemann et al., 2014), including Minoan terranes (Zulauf et al., 2007; Zlatkin et al., 2013), was traditionally attributed to distant dispersion by large river systems (Zeh et al., 2001), and recently to a super-fan system shedding detritus with ca, 1.0 Ga-old zircons from the Transgondwanan supermountain towards the Gondwana margin (Meinhold et al., 2013). Minor quantities of 1.6–2 and 2.1–2.5 Ga zircon ages determined in this study (Fig. 12), could potentially be attributed to sources from Arabian–Nubian shield (e.g. Stacey and Hedge, 1984; Ali et al., 2009), West African (Nance and Murphy, 1994), and/or South-China craton (Zhang and Zheng, 2013). In conclusion, the basement of the SMM was most probably located in eastern parts of the north Gondwana margin in late Neoproterozoic, i.e. northeast of Saharan metacraton and southwest of South-China craton.

5.6. Tectonic evolution

From late Ediacaran to mid-Cambrian (562–522 Ma), the SMM together with the basement of the Struma Unit represented a part of an Andean-type continental magmatic arc situated along the northern margin of Gondwana (Fig. 13a). Based on U–Pb zircon age record, similar late Neoproterozic position could also be suggested for the Pirgadikia micro-terrane of the Internal Hellenides, Lotru unit of the Carpathians and the İstanbul zone of the western Pontides (Fig. 12). Although xenocryst ages, Lu/Hf trends and DM model ages of zircons from the magmatic rocks in the SMM suggest reworking of an older continental basement in which they were intruded (Fig. 10), these continental basement rocks were not recognised in the study area. However, such rocks were reported in the neighbouring Tran region in the Struma Unit (Kounov et al., 2012), in basement from the Pelagonian zone (Anders et al., 2006; Schenker et al., 2014; Zlatkin et al., 2014), and the İstanbul (Ustaömer et al., 2005) and the Sakarya zones (Aysal et al., 2012) in northwestern Turkey. This hypothetical continental basement was adjacent to an Ediacaran accretionary wedge or a fore-arc basin dominated by terrigenous sediments, presently a part of the Lower Complex (Dimitrijević, 1967; Krstić and Karamata, 1992), whereas the pre-Ordovician Vlasina Unit would correspond to an external part of the wedge dominated by sediments scraped off the ocean bottom (Fig. 13a; Milovanović et al., 1988; Popović, 1991 and references therein). Both the terrigenous and ocean-floor-dominated parts were subsequently intruded by magmatic rocks of the late Cadomian arc. This magmatic arc was most probably related to the oblique subduction of the Prototethys (Fig. 13a), with dextral transform boundary already terminating subduction in the western (i.e. Avalonian) section of the belt (Nance et al., 2002, 2010). Given the proximity of the trench and the oblique direction of the subduction, a minor intra-oceanic thrusting might have been initiated, which potentially led to a partial obduction of small oceanic fragments represented by the ophiolites of Frolosh Unit, on a still active continental magmatic arc (Fig. 13b). A "gap" in the subducting slab due to the missing obducted segment would result in incursion of depleted mantle into the mantle wedge. Increased heat flow would promote partial melting at the base of the arc resulting in the formation of granites and gabbros with zircons crystallised in a juvenile melt contaminated with crustal material (Figs. 10b and 13b; Lisina gabbro, Vlajna and Doganica granites). Units with similar age of magmatic-arc activity and basement provenance, as Sebeş–Lotru in the Carpathians or Pirgadikia Unit in the internal Hellenides are devoid of ophiolitic segments and mixed magmatic sources, suggesting minor size and local importance of this obduction. Increased load on the margin of the magmatic arc could cause slight subsidence of the entire arc facilitating deposition in a retro-arc basin, currently represented by para-metamorphic series of the Lower Complex (Fig. 13b). The continuation of the subduction of the Prototethys in the early Cambrian reinitialised continental arc magmatism in Vlasina Unit (granite from the Božica magmatic complex), and the basement of Struma Unit (i.e. COL complex including Bosilegrad monzonite, and SDC including Delčevo granite; Fig. 13c). Termination of the subduction and transition to sheared continental margin via dextral wrenching reported for Avalonia between 605 and 545 Ma (Nance et al., 2002), and central Cadomia at ca. 540 Ma (Linnemann et al., 2007), potentially took place in the studied region after 521 Ma. Alternatively, termination of activity along this margin occurred by a southward-migrating intra-oceanic subduction zone (Stampfli et al., 2013).

Following the period of magmatic quiescence, predominantly peraluminous, leucocratic granitic magmatism indicates a reactivation of the North-Gondwanan active margin (i.e. Vinica dyke, Kukavica and Bukovac granites, and Maleševski Mts. orthogneiss Fig. 13d). This felsic magmatism is probably caused by the subduction of the Qaidam back-arc ocean or the remaining Protothetyan domain (Stampfli et al., 2013). Lower Ordovician sediments were deposited in the Vlasina Unit (i.e. post-Cambrian Vlasina), and in the Veles series (Pavlović, 1962; Karamata and Krstić, 1996). The former could represent a fore-arc basin, while the sediments of the latter were most probably deposited in the retro-arc or an incipient back-arc basin (Fig. 13d).

Widespread within-plate mafic magmatism occurred in the SMM in late Middle Ordovician (456–462 Ma; Vinica and Golemo Selo amphibolites; Fig. 13d). Most of these tholeiitic dykes were intruded in the Lower Complex, whereas the variety of geochemical signatures of the mafic magmatic rocks in the Vlasina Unit suggests that they represent a collection of various sources, probably amalgamated during the earlier, subduction-related accretion of this unit. The basaltic magmatism from the Lower Complex could possibly be attributed to the rifting of a back arc basin (Fig. 13e), although felsic igneous rocks of this age are absent in the study area, and marine sedimentation, including carbonates, continued uninterrupted on both flanks of the SMM (Pavlović, 1977; Karamata and Krstić, 1996; Banjac, 2004). Remnants of these sediments are presently part of the Eastern Veles series and Vlasina Unit. Arc-related magmatism was reported southeast of the study area (in present coordinates; i.e. Ograzhden block), from Middle Ordovician to early Silurian (Macheva et al., 2006; Zagorchev et al., 2011a; Kounov et al., 2012). Similar magmatic-arc activity was determined in the Cumpăna Unit (Balintoni et al., 2010a), and the Carpathian-type units in Romania (Balintoni and Balica, 2013b), as well as the Vertiskos Unit in Greece (Himmerkus et al., 2009a; Meinhold et al., 2010). Since the host rocks of these magmatic bodies are upper Cambrian to Lower Ordovician sediments, we suggest

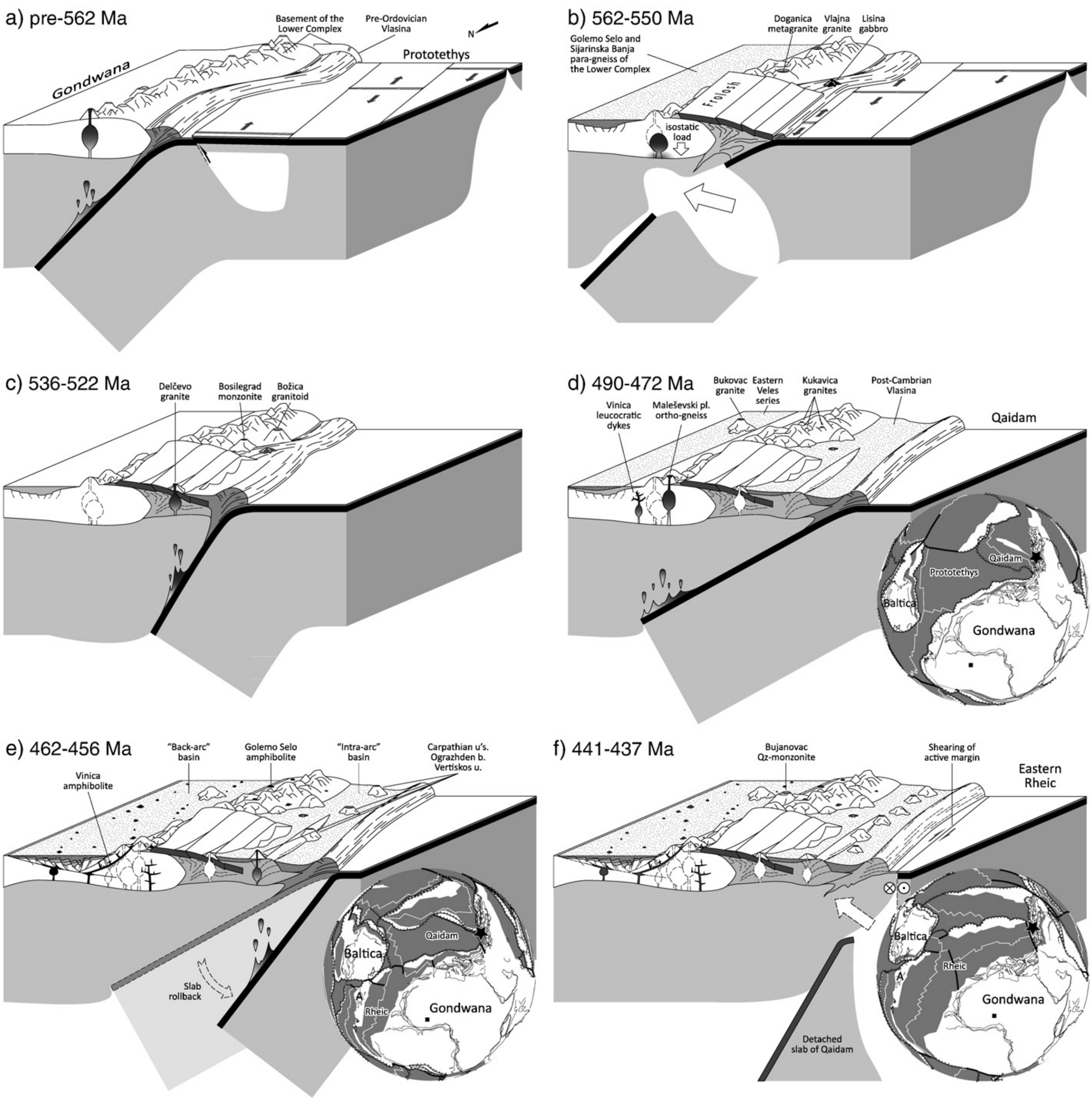


Fig. 13. Unscaled sketches of the inferred tectonic evolution of the study area since the Cadomian until the earliest Mesozoic. Dot-stippled areas represent basins with ongoing sedimentation (both clastic and biochemical), and black units designate emplacement of mafic and ultramafic rocks. In (d), (e), (f), and (g), the location of the SMM is designated by a black star on the global reconstruction of Stampfli et al. (2013), where black square represents the South Pole, white line a spreading centre, thick black line a wrenching zone, and notched black line an active margin. See text for details.

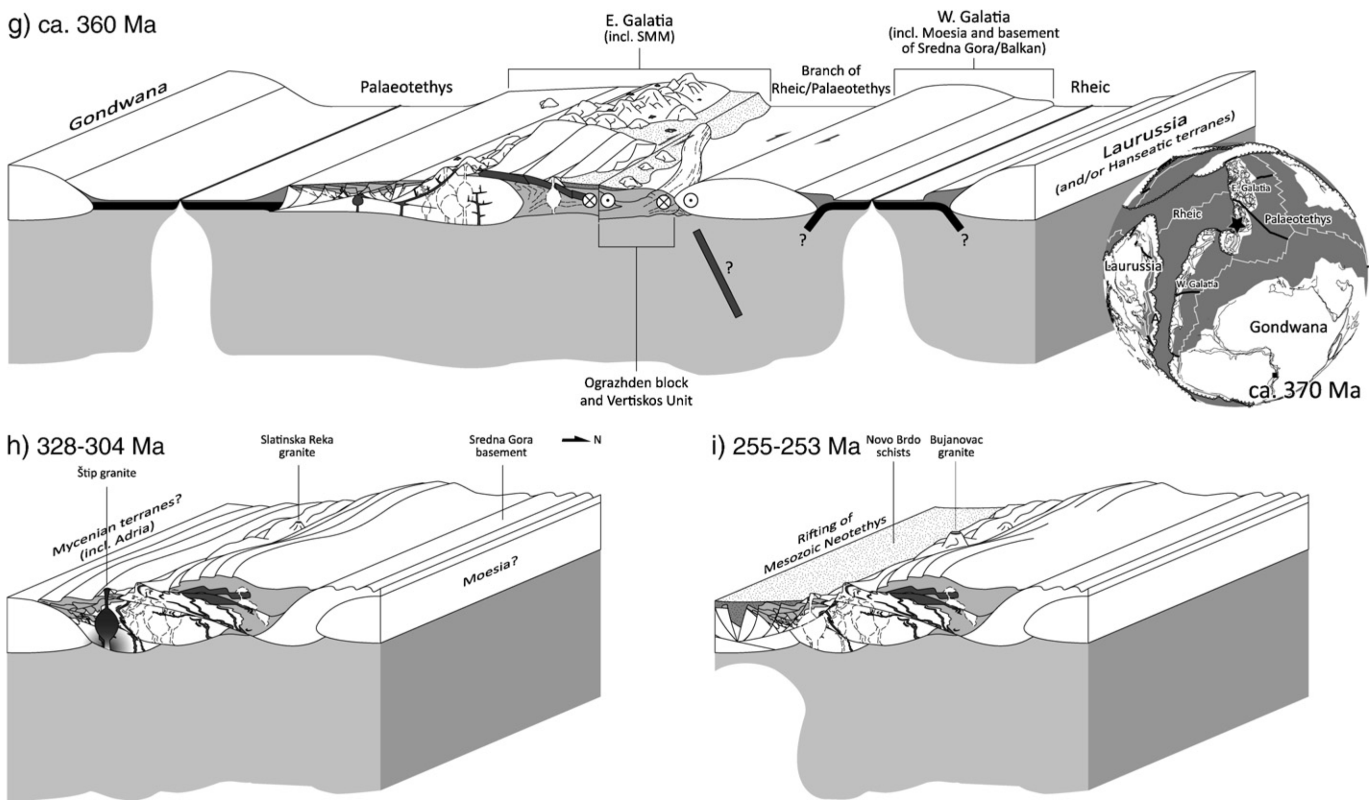


Fig. 13 (*continued*).

oceanward migration of the magmatic arc due to slab rollback, with continuing passive-margin deposition in thus formed "intra-arc basin" (i.e. post-Cambrian Vlasina; Fig. 13e).

Subduction was terminated in the early Silurian by dextral shearing of the subducting slab, related to an oblique opening of the Rheic Ocean (Fig. 13f; Stampfli et al., 2013). Within-plate peraluminous Bujanovac Qz-monzonite is possibly a result of the partial melting of the continental crust due to high thermal gradient resulting from the penetration of the astenospheric material into the mantle wedge following the detachment of the sheared slab (Fig. 13f, 439 ± 2 Ma). Magmatism in the Vertiskos Unit in Greece (Himmerkus et al., 2009a; Meinhold et al., 2010), and Ograzhden block in Bulgaria (Zagorchev et al., 2011a), that lasted until 426 and 433 Ma respectively, shows a predominant magmatic-arc affinity. Since the magma sources in these two areas were found to be heavily contaminated with crustal material, there is a distinct possibility that this magmatism was also caused by the process proposed for the formation of the Bujanovac Qz-monzonite.

During the subsequent period of magmatic quiescence, the SMM had presumably drifted westwards across the Rheic Ocean towards Laurussia, as an eastern part of the ribbon-like Galatian super-terrane (Karamata, 2006; Stampfli et al., 2013). Meanwhile, Eastern Galatia had collided with its western counterpart generating dextral wrenching of constituent units and opening of pull-apart basins (Fig. 13g; Stampfli et al., 2013). This event resulted in eastward movement of Ograzhden block and Vertiskos Unit, and concomitant deposition of Upper Devonian turbidites with evidence of mafic volcanism along the eastern part of the Vlasina Unit (Spassov, 1973; Zagorčev and Bončeva, 1988; Krstić et al., 2002). This depositional setting persisted until the lower Carboniferous when the final amalgamation/juxtaposition of the SMM to the Western Galatia (including Moesia and possibly also basement of Balkan and Sredna Gora units), most probably took place (Haydoutov and Yanev, 1997; Banjac, 2004 and references therein; Karamata, 2006; Gerdjikov et al., 2010). Subduction direction of the branch of Rheic or Palaeotethys ocean that was located between the two parts of the Galatian super-terrane is still uncertain (e.g. Iancu et al., 2006; Plissart, 2012). Sustained collision of segments of the Galatian super-terrane and the margin of Laurussia had probably caused top-to-the-west thrusting along the SMM (in present orientation), separating it into distinct structural levels (Fig. 13h). An overall crustal thickening would have promoted metamorphism, varying according to depth at which each of these segments was buried. Ultimately, this tectonic over-burden could lead to the partial melting of the lower crust, possibly resulting in emplacement of the minor granitic bodies in Vrvi Kobila area (i.e. Slatinska Reka granite, 328 ± 5 Ma), and a late peraluminous granite within the Štip magmatic complex in the Eastern Veles series (304 ± 3 Ma, Fig. 13h). Similar magmatism was not found in the rest of the SMM, but was reported further east in the basement of Sredna Gora and Balkan units (Carrigan et al., 2005), and the central Rhodope complex (Peytcheva et al., 2004). The southern boundary of the SMM in late Carboniferous was either an active northward subduction zone of Palaeotethys (Zulauf et al., 2015), or it was occupied by the Mycenian terranes which were also part of the Eastern Galatia, juxtaposed to the SMM by dextral wrenching (Stampfli et al., 2013; Fig. 13h).

The final magmatic episode along the SMM is represented by the emplacement of the late Permian–Early Triassic peraluminous Bujanovac granites (SM199-1 and SM377-1, ca. 255 Ma, Fig. 13i). Similar igneous activity reported elsewhere along the European continental margin was related to the closure of the remaining Palaeotethyan domains and the following extension which consequently led to the opening of the Mesozoic Tethys (Robertson and Karamata, 1994; Karamata and Krstić, 1996; Pe-Piper, 1998; Stampfli et al., 2002; Karamata, 2006; Himmerkus et al., 2009b; Burg, 2012).

6. Conclusions

In the present study, the geochemical analysis, Hf isotope measurements, and U–Pb LA-ICP-MS zircon dating of igneous and sedimentary rocks from the Serbo-Macedonian Massif and the basement of the adjacent units have allowed the following conclusions to be made:

– During the Cadomian orogenic cycle the Lower Complex of the SMM was part of a continental magmatic arc developed along the northern margin of Gondwana. The complex could be correlated with the Lotru and Pirgadikia units of the Carpathians and the Internal Hellenides, respectively. Based on inherited and detrital zircon data, its late Neoproterozoic position in the eastern part of the northern margin of Gondwana is tentatively proposed (i.e. northeast of Saharan metacraton and southwest of South-China craton).
– The volcano-sedimentary pre-Ordovician Vlasina Unit most probably represents part of a late Neoproterozoic accretionary wedge dominated by ocean-floor sediments. During this time the Lower Complex was most probably a more internal part of the same volcano-sedimentary succession or a fore-arc basin.
– The magmatic-arc activity in the study area was detected in the Ediacaran to the mid-Cambrian in the Lower Complex, Vlasina Unit and the basement of Struma Unit. These units together with the Eastern Veles series, experienced renewal of arc-related magmatism in the late Cambrian to Early Ordovician, while mafic magmatism related to rifting occurred in the Lower Complex during Middle to Late Ordovician. Minor pulse of within-plate felsic magmatism in the Lower Complex took place in the early Silurian.
– The age of the high-strain deformation and peak metamorphism in the SMM was constrained to the Variscan orogeny by the presence of highly deformed early Silurian meta-granites and undeformed late Carboniferous granites intruding the gneisses of the Lower Complex.
– The Variscan collisional tectonics led to the differential burial of constituents of the SMM (i.e. Lower Complex, Vlasina Unit, basement of Struma Unit, and Eastern Veles series), resulting in different metamorphic grades of these domains with a common late Neoproterozoic–early Palaeozoic history. Additionally, this complex Variscan framework was subsequently severely disrupted by the Alpine events.
– During the Permian–Triassic the entire SMM experienced widespread crustal extension and emplacement of within-plate granites, most probably related to the rifting of the Mesozoic Neotethys.

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