LATE VALANGINIAN-HAUTERIVIAN RADIOLARIAN AGE CONSTRAINTS FOR SUBDUCTION-RELATED SUBMARINE VOLCANIC ACTIVITY IN THE AMASIA-STEPANAVAN OPHIOLITE (NORTHERN ARMENIA)

Ghazar Galoyan*, Taniel Danelian,✉, Gabriel Ojo**, Sylvie Regnier**, Lusine Atayan*, Sona Amiraghyan*** and **Arayik Grigoryan***

* *Institute of Geological Sciences, National Academy of Sciences of Republic of Armenia, Yerevan, Armenia.*

** *Univ. Lille, CNRS - UMR 8198, France.*

✉ *Corresponding author, email: taniel.danelian@univ-lille.fr*

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ABSTRACT

Radiolarian biochronology of radiolarites associated with ophiolitic lavas in northern Armenia provides important age constraints for the geodynamic reconstruction of Tethys in an area that makes the link between the Lesser Caucasus and the Eastern Pontides. We present results obtained from a moderately well-preserved radiolarian assemblage extracted from radiolarian cherts that crop out in the Bazoum Horst (between Stepanavan and Amasia). The age of these radiolarites may be restricted to the late Valangingian-Hauterivian based on the co-occurrence of the radiolarian species *Archaeodictyomitra pseudoscalaris* (Tan), *Crolanium bipodium* (Parona), *Pseudodictyomitra suyarii* Dumitrica and *Tethysetta ovoidala* Dumitrica. The dated radiolarites lie stratigraphically ca. 7-8 m above basaltic lavas that have a subduction-related geochemical signature. In combination with results from the Amasia ophiolite, this indicates the presence of a subduction-related island arc at the scale of the Amasia-Stepanavan oceanic realm that was active during Valanginian to Barremian times.

INTRODUCTION

Mesozoic radiolarians are particularly useful for establishing a timeline for Tethyan rock sequences, especially radiolarites, which are often associated with the emplacement of submarine lavas. Therefore, radiolarian dating of these siliceous biogenic sequences, which are usually devoid of any other fossils, is of great significance for the reconstruction of the geodynamic and paleoenvironmental evolution of past oceanic domains that are often preserved in tectonically complex areas (e.g., Al-Riyami et al., 2002; Vrielynck et al., 2003; Danelian et al., 2006; 2008a; Goričan et al., 2012; Ferrière et al., 2015; Avagyan et al., 2017; Varnava et al., 2021; Okay et al., 2022; Cordey, 2022; Chiari et al., 2023; Naing et al., 2023; Sashida et al., 2023).

Knowing the age of radiolarites that occur in the ophiolitic zones of Armenia is thus particularly important for the reconstruction of the geodynamic history of the Tethyan realm preserved in the Lesser Caucasus. Initial radiolarian studies were conducted at the time of the Soviet Union (Vishnevskaya, 1995; Knipper et al., 1997), but most of the radiolarian ages were obtained during the last 20 years and in the framework of the Armenian-French collaboration on the Tethyan geology of the Lesser Caucasus (e.g., Danelian et al., 2007; 2008b; 2010; 2012; 2014; 2016; 2017; 2023; Asatryan, 2009, Asatryan et al., 2010; 2011; 2012).

The Armenian ophiolites are part of an Alpine orogenic belt, which starts in northern Iran and continues through Armenia to northeastern Turkey (e.g., Aslanyan and Satian, 1977; Knipper, 1980; Adamia et al., 1981; Zakariadze et al., 1983; Galoyan et al., 2007; 2009; Galoyan, 2008; Rolland et al., 2009a; 2010; Sosson et al., 2010; 2016). As such, several Tethyan geodynamic analyses of the region took a parallel look at ophiolites and related sequences in both Eastern Turkey and NW Armenia (Rolland et al., 2009b; Robertson et al., 2014; Rolland, 2017), considering them as the relics of a 700-km-long single ophiolitic nappe (Hässig et al., 2013a, 2013b). The Amasia - Stepanavan ophiolites, situated in north-western Armenia, have a particular significance in this respect, as they make the link between the ophiolite zones running through Eastern Armenia and northeastern Turkey. In this work, we present radiolarian biostratigraphic results from radiolarites overlying basaltic lavas at the Bazoum Horst, which is part of the wider Amasia-Stepanavan ophiolites in NW Armenia.

REGIONAL GEOLOGY

The ophiolites of the Lesser Caucasus are part of a Tethyan suture zone (e.g., Knipper, 1980; Adamia et al., 1981; Zakariadze et al., 1983; Knipper et al., 1986; Dercourt et al., 1986; Galoyan, 2008; Sosson et al., 2010 and references therein). They represent the relics of a Mesozoic oceanic realm that was situated between Eurasia and the South Armenian Block (SAB; Dercourt et al., 1986; Kazmin et al., 1987; Sosson et al., 2010; Galoyan et al., 2020), a microcontinent that is mainly characterized by its Upper Devonian-Permian sedimentary sequences (see Arakelyan, 1964) and was detached from Gondwana during the Triassic (see Nikogosian et al., 2023 and references therein). To the North, the Eurasian margin (or locally the Transcaucasian Massif) is regarded, in general, as an active continental margin for most of the Mesozoic (e.g., Kazmin et al., 1987). It is also known as the Somkheto-Karabagh Belt (or terrane) and is mainly defined by its widespread Middle Jurassic to Upper Cretaceous volcano-sedimentary sequences (e.g., Adamia et al., 1981; Sosson et al., 2010; Mederer et al., 2014; Rolland, 2017 and references therein). Alternatively, the Somkheto-Karabagh terrane is regarded by Galoyan et al. (2018) as an island arc (and not as an active continental margin). The obduction of the Mesozoic oceanic crust onto the South

Armenian Block took place later, during the Turonian to Coniacian time interval (Sokolov, 1977; Sosson et al., 2010; Hässig et al., 2013a; 2013b; Danelian et al., 2014; 2023; Rolland et al., 2020).

The ophiolite outcrops in the Lesser Caucasus mainly occur in two zones (Fig. 1).

The *Sevan-Hakari (Akera) ophiolite zone* is mainly known after the work of Palandjyan (1971), Aslanyan and Satian (1977), Knipper (1980), Knipper and Khain (1980), Galoyan (2008) and Galoyan et al. (2009). The ophiolite is exposed E and SE of the Lake Sevan, as well as in the territory of Artsakh (Mountainous Karabagh) and represents the main Tethyan suture zone in the Lesser Caucasus (e.g., Sosson et al., 2010). Smaller, but distinct ophiolitic outcrops, situated in northern Armenia (i.e. the Amasia and Stepanavan ophiolites) are regarded as the north-western extension of the Sevan-Hakari ophiolite zone (Galoyan et al., 2007; Galoyan, 2008; Rolland et al., 2009a, 2010; Sosson et al., 2010; Galoyan and Melkonyan, 2011; Hässig et al., 2013a; 2013b; 2019 and references therein), forming a ca. 400 km-long ophiolite belt running from Amasia (NW Armenia) to the Araks Valley (southern Karabagh).

The *Vedi ophiolite*, located to the southwest of Lake Sevan and about 45 km southeast of the city of Yerevan, is mainly known from outcrops around and within the Khosrov natural reserve of the Vedi area; it corresponds to a folded klippe structure (Sokolov, 1977; Knipper and Sokolov, 1977; Sosson et al., 2010). Based on borehole data, this ophiolite unit is also present westwards, but buried under 1-5 km-thick Mesozoic-Cenozoic sedimentary sequences and the Quaternary sediments of the Araks Valley (for more details see Galoyan, 2008).

According to several studies published in the framework of the French-Armenian collaboration (e.g., Galoyan et al., 2007; 2009; Sosson et al., 2010; Hässig et al., 2019, and references therein) the Amasia-Stepanavan-Sevan-Akera ophiolites were formed in a supra-subduction intra- to back-arc setting, north of a north-dipping intra-oceanic subduction zone that was active at least since the Toarcian. Alternatively, for Galoyan et al. (2018), the Somkheto-Karabagh terrane represents an island arc that was formed during the Jurassic above a southward-dipping subduction zone within the Paleotethys and the ophiolites of the Lesser Caucasus originated south of the Somkheto-Karabagh island arc. Finally, Nikogosian et al. (2023) proposed that the Lesser Caucasus ophiolites were formed in a fore-arc setting, at a north-dipping subduction zone that operated underneath the Somkheto-Karabagh block (or Transcaucasian plate).

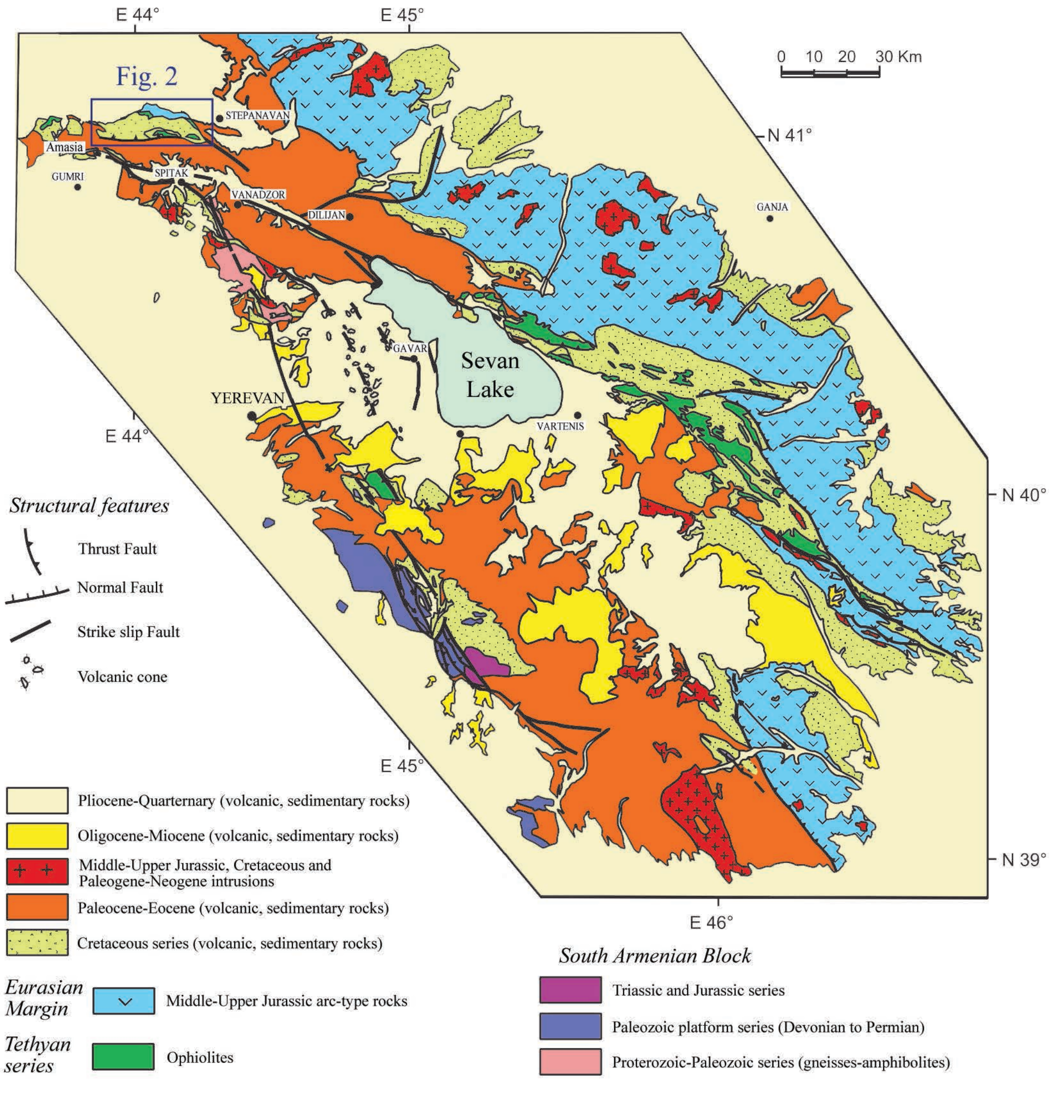

Fig. 1 - Geological map of the Lesser Caucasus (modified after Sosson et al., 2010), including the location of the studied area shown in a frame (Fig. 2).

GEOLOGICAL SETTING OF THE STUDY AREA

The Bazoum Horst (or horst-anticlinorium) is a 10 km-wide and 35 km-long structure that is situated in the western part of the Bazoum mountain range of Northern Armenia (Figs. 1 and 2). It corresponds to a highly folded structure with a complex multi-stage magmatic, tectonic, and metamorphic history. The "Stepanavan (or Gargar) ophiolite" (Galoyan et al., 2007) is part of this Horst and represents only its smaller, eastern segment. Within the Horst many exposures of ophiolitic rocks are part of the Amasia-Stepanavan branch of the NW Armenian ophiolites. Accordingly, these are considered as the northwestern extension of the Sevan-Akera ophiolite zone (e.g., Palandjyan, 1975; Aghamalyan, 1998; 2007) or the Amasia-Sevan-Hakari suture zone (e.g., Galoyan, 2008; Galoyan and Melkonyan, 2011; Hässig et al., 2013a; 2013b).

Located between the towns of Stepanavan and Amasia, the Bazoum Horst has been known since the 1960s as the Bazoum Uplift, or Horst-anticlinorium. It represents the oldest lithologies within an "anticlinorium" cropping out in the Sevan-Shirak synclinorium that was formed during the Paleogene (Sarkisyan, 1966). Alike for the Sevan ophiolite, there is no evidence within the boundaries of this complex structure for their obduction onto an older, autochthonous formation, such as the SAB. The Horst is bound both to the north and the south by large deep faults, which are inherited since the Cretaceous (?) and reactivated many times afterwards (e.g., Gabrielyan et al., 1968; Palandjyan, 1975). According to these authors, the Horst-anticlinorium is limited to the east by a third fault, the Pushkin's Revers fault.

The metamorphic rocks in the area are considered to be either of Precambrian-Early Paleozoic (Barkanov, 1937; Paffenholtz, 1946; 1959) or Late Jurassic-Cretaceous age (Aslanyan, 1958). They were first described in detail by Palandjyan (1975) and Aghamalyan (1978 and references therein); they are represented by greenschists and blueschists, amphibolites, phyllites, marbles. Late Cretaceous ages (90-80 Ma) were obtained on glaucophane schists using the K-Ar method (Baghdasaryan et al., 1962), which were later confirmed by ^{40}Ar-^{39}Ar dating (95-90 Ma; see Rolland et al., 2009a for details). Within the Horst, serpentinized peridotites, serpentinites, and minor gabroids are the main plutonic part of the ophiolites (e.g., Galoyan, 2024 and references therein), which until the 1970s were considered to be extensional "intrusions" or "large dykes" into the Mesozoic-Cenozoic formations. The coexistence of these mafic-ultramafic rocks with the "basalt-radiolarite" series, as a single ophiolite complex, was first reported by Palandjyan (1975). Later on, in the framework of the Armenian-French collaboration, the Stepanavan ophiolites were studied in further detail (Galoyan et al., 2007; Danelian et al., 2007; Rolland et al., 2009b; Hässig et al., 2019). During the last two years, detailed fieldwork allowed the discovery of large exposures of basaltic lavas, including radiolarites, which were omitted in previous studies and maps as "ophiolites" (Fig. 2). Moreover, the nature of the southern contact of the Horst or of the ophiolites was recently documented and discussed (Galoyan, 2024). The Cretaceous sedimentary formations in the area have been thoroughly studied by Hakobyan (1962), who distinguished five Lower Cretaceous sedimentary sequences (or "suites") and an Upper Cretaceous one. The contacts between these sequences and the ophiolite complex are mainly tectonic, with the exception of the Upper Cretaceous sequence, which is sometimes clearly transgressive on ophiolitic and other lithologies.

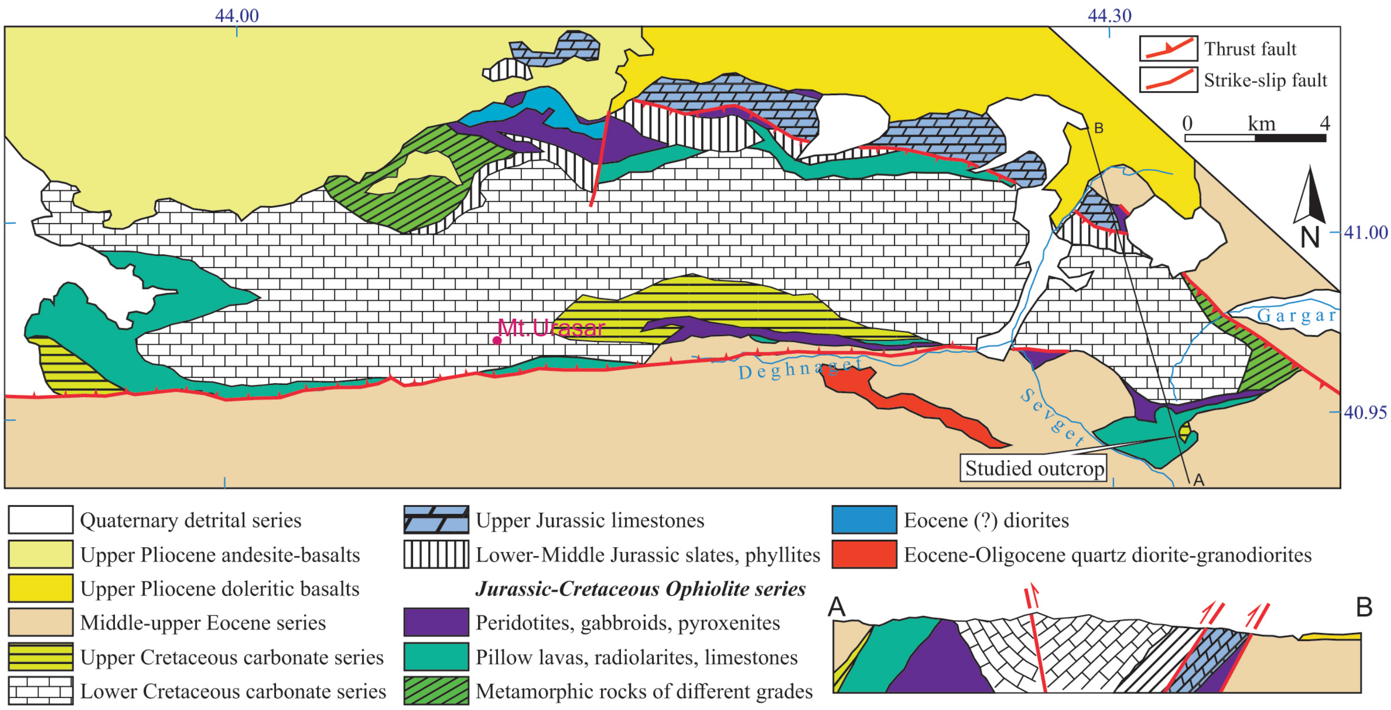

Fig. 2 - Schematic geological map of the Bazoum Horst, compiled with the data of our own field and laboratory research and supplemented with the data of former maps (Hakobyan, 1962; Palandjyan, 1975).

MATERIAL AND METHODS

Fieldwork relevant to this study was conducted during the years 2011; 2014; 2022 and 2023; this investigation confirmed that both aphyric and porphyritic basalts and basaltic andesites are sometimes overlain by reddish-gray radiolarites and siliceous limestones rich in foraminifera. These lavas often form pillows and are part of the ophiolitic complex.

In order to date the radiolarites cropping out in the Bazoum Horst an extensive sampling of the chert sequences was carried out. However, radiolarians were observed only in six samples, in thin-section preparations. After hydrofluoric acid leaching of all six samples at the University of Lille, only one out of these six samples yielded abundant, moderately well-preserved and identifiable radiolarians.

The studied section (Figs. 2 and 3a) is exposed on the right bank of a small ravine close to the watershed of the Gargar and Sevget rivers (N 40.94531°, E 44.32325°). Here, an over 10 m-thick sequence of dark reddish ribbon radiolarites stratigraphically covers basaltic lavas, which are devoid of any clear pillow appearance. The radiolarian-bearing sample (St11.13) was collected 7-8 m above the underlying lavas. Laterally, ca. 50 m to the east of the studied outcrop, pink and gray limestones, of likely Late Cretaceous age (Hakobyan, 1962) occur. In thin section observations, the radiolarian-bearing sample (St11.13) displays numerous radiolarians with well-distinct test outlines (Fig. 3b). The chert samples were processed with dilute hydrofluoric acid (HF 5%) at the Evo-Eco-Paléo laboratory of the University of Lille. Radiolarians were extracted after repetitive HF attack for ca. 24 hours; the microfossils were picked up from dry residues obtained after each leaching; the promising specimens were mounted on SEM stubs and after gold coating, they were photographed with the help of a Zeiss EVO-MA10 scanning electron microscope. Taxonomic concepts applied during this study follow mainly those of Baumgartner et al. (1995a), O'Dogherty (1994), and Dumitrica et al. (1997; 2022).

Thin section preparations and geochemical analyses were conducted at the Institute of Geological Sciences, at Yerevan. Here the major element analyses were obtained after quantitative chemical analysis ("HCAM 138-X" Chemical methods; for details see https://meganorm.ru/Index2/1/4293753/4293753734.htm, in Russian), which is an accelerated method to determine rock-forming elements in rocks and ores.

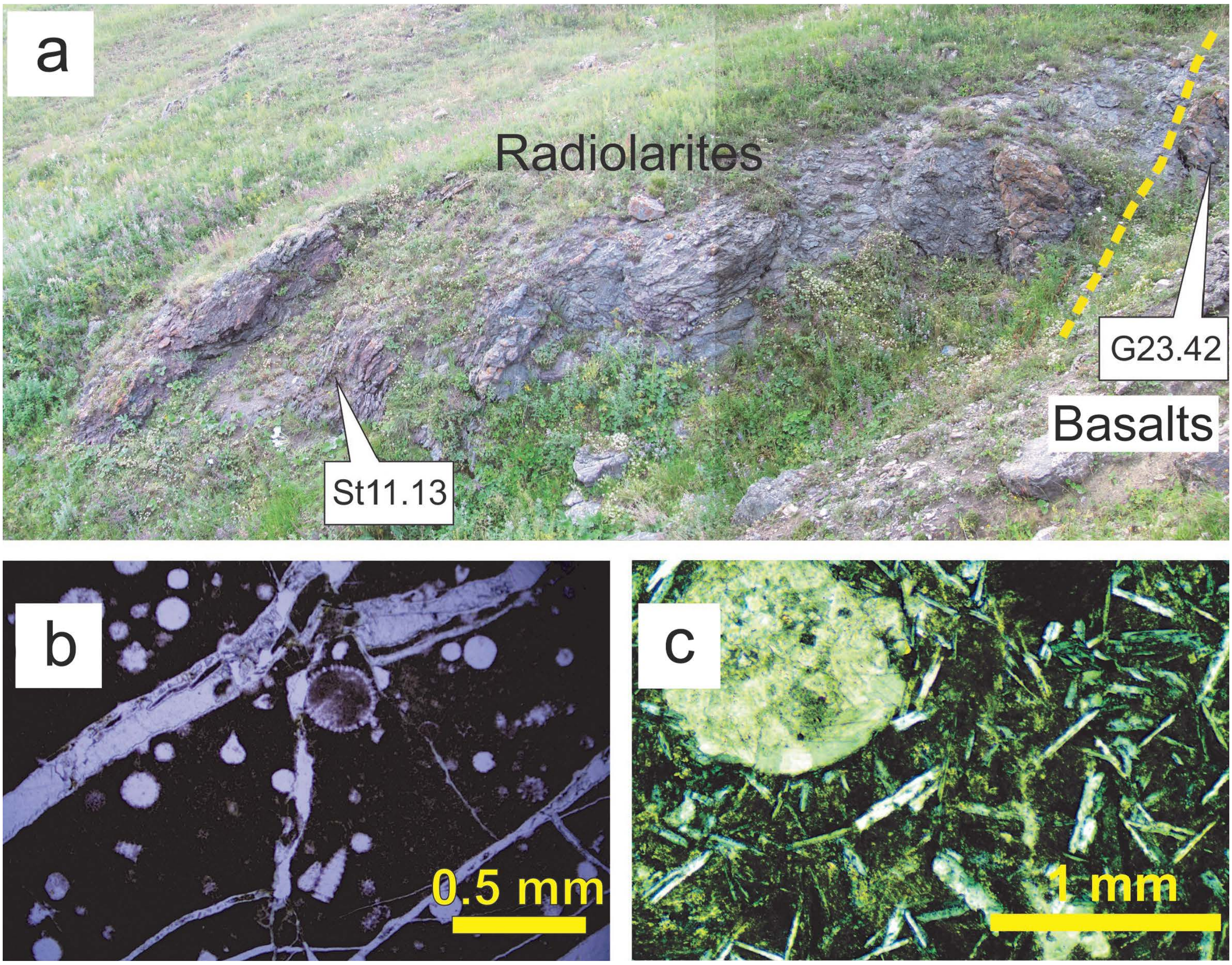

Fig. 3 - a) Field view of the studied outcrop displaying a ca. 10 m-thick radiolarian chert sequence (on the left) overlying basalts (on the right); b) thin section microphotograph of the studied radiolarian chert sample St11.13; c) thin section microphotograph of the studied lava sample G23.42).

RESULTS

Radiolarian assemblage and age

The radiolarian species identified are illustrated in Fig. 4. According to the biozonation of Baumgartner et al. (1995b) the radiolarian assemblage identified in the sample St11.13 may be assigned to the Unitary Association Zones (UAZs) 17-22 and correlated with the late Valanginian to late Barremian/early Aptian interval. This biozone and age assignment is based on the co-occurrence of *Archaeodictyomitra pseudoscalaris* (Tan) sensu Baumgartner et al. (1995a) and of *Crolanium bipodium* (Parona) *sensu* Dumitrica et al. (1997) (= *Crolanium pythiae* Schaaf in Baumgartner et al., 1995a), two species known only to first occur since the late Valanginian UAZ 17 and a number of long ranging species that are still present in the youngest UAZ 22 e.g., *Acaeniotyle umbilicata* (Rüst), *Angulobracchia portmanni* Baumgartner, *Archaeodictyomitra pseudoscalaris* (Tan), *Crolanium bipodium* (Parona) *sensu* Dumitrica et al. (1997) (= *Crolanium pythiae* Schaaf in Baumgartner et al., 1995a), *Pantanellium squinaboli* (Tan) and *Stylosphaera* (?) *macroxiphus* (Rüst).

Moreover, according to the biozonation of O'Dogherty (1994), *A. umbilicata* and *A. portmanni* last occur in his UA 9 (*costata* subzone), which is correlated with the middle Aptian/early Albian age interval.

In addition, *Pseudodictyomitra suyarii* Dumitrica is only known between the late Valanginian and the early Aptian age interval (Dumitrica et al., 1997; Andjic et al., 2019), while *Novixitus robustus* (Wu) *sensu* Dumitrica et al. (2022) ranges between the Valanginian and the Aptian and *Archaeodictyomitra mitra* Dumitrica between the Valanginian and the late Hauterivian-early Aptian as reported by Dumitrica et al. (2022). Moreover, according to O'Dogherty (1994), *Crolanium bipodium* (Parona) last occurs in the late Barremian *Asseni* zone (UA 1) of his biozonation. Finally, Dumitrica et al. (2022) refined the species concept of *Tethysetta ovoidala* Dumitrica and reported that its age range covers the latest Tithonian to Hauterivian age interval. In conclusion, the age of sample St11.13 may be restricted to the late Valanginian - Hauterivian interval based on the co-occurrence of *Archaeodictyomitra pseudoscalaris*, *Crolanium bipodium*, *Pseudodictyomitra suyarii* and *Tethysetta ovoidala*.

Petrology and geochemistry of lavas

A basalt sample collected from the volcanic sequence overlain by the dated radiolarites (G23.42, Fig. 3a) was studied in detail. The volcanic rocks are characterized by amygdaloidal structure and aphyric texture (Fig. 3c). Irregularly oriented microlites and needles of plagioclase are included in the former hyalopilitic main matrix, which is now extensively recrystallized and transformed. It is mainly filled with quartz-chlorite and mostly carbonate material. Micropores (~ 0.5 mm, amygdule) and cracks are frequent in the rock, which are also filled with the above-mentioned new minerals. Major element composition of the sample G23.42 is reported in Table 1.

Based on its projection in the TAS (Total Alkali-Silica) diagram (Fig. 5a), the analyzed sample G23.42 is a trachybasalt. However, the widespread occurrence of secondary minerals observed in thin section suggests that this classification must be considered with some caution as Na_2O and K_2O are highly mobile during seawater alteration. Overall, the sample is characterized by a subduction-related geochemical signature, as indicated by the tectono-magmatic discrimination diagrams of its less mobile major elements. In particular its composition falls in the "Island-arc and active continental margin basalts" and "Island-arc tholeiites" fields as defined, respectively, by FeO-MgO-Al_2O_3 and TiO_2-MnO-P_2O_5 diagrams (Fig. 5b-c).

DISCUSSION

Fig. 6 presents the results of this study integrated in the recent synthesis made by Danelian et al. (2023) for all dated radiolarites in the ophiolitic units of the Lesser Caucasus. Regarding the Amasia-Stepanavan ophiolite, the oldest radiolarites found to date are of Late Jurassic age. Indeed, Danelian et al. (2016) dated middle Oxfordian - lower Kimmeridgian and Berriasian radiolarites overlying stratigraphically ophiolitic lavas in the Amasia area. Near Stepanavan (in the Gargar Valley), east of the outcrop of this study, Danelian et al. (2007) dated upper Kimmeridgian - lower Tithonian radiolarites stratigraphically covering ophiolitic lavas. The youngest radiolarites stratigraphically overlying submarine lavas are dated as Cenomanian (Danelian et al., 2014). They occur near the town of Amasia and close to a particularly interesting outcrop of over 15 m-thick radiolarian-rich pyroclastic sequence dated as late Barremian in age (Danelian et al., 2016). Although no trace element geochemical evidence exists so far for all these radiolarite-associated lavas, the volcanic rocks of presumable Late Cretaceous age described from the eastern part of the studied region (namely, the Sevget Valley area) have a subduction-related geochemical signature (Galoyan et al., 2007). It is noteworthy that unlike the lava sample analyzed in this study (G23.42), most of those volcanic rocks display a large porphyritic texture. The subduction setting is consistent with the presence of blueschist facies rocks metamorphosed during the Cenomanian-Turonian (~ 95-90 Ma), before their subsequent exhumation during the late Campanian-early Maastrichtian (~ 73-71 Ma, Rolland et al., 2009a).

Combined with the radiolarian-rich submarine pyroclastic rocks dated as late Barremian in age (Danelian et al., 2016), our results from the Bazoum Horst allow us to reconstruct a more coherent story for the Tethyan oceanic realm preserved between Amasia and Stepanavan. We may now consider that the volcanic activity in this part of Tethys was influenced by subduction-related processes for at least the Valanginian to Barremian interval, which should be considered as a minimal age range, given the subalkaline/alkaline submarine lavas of latest Tithonian/Berriasian age recently established for the Sevan ophiolite (Seyler et al. 2023; Danelian et al. 2023).

Table 1 - Major element (wt. %) whole-rock analysis of the sample G23.42.

SiO_2	Al_2O_3	TiO_2	Fe_2O_3	FeO	CaO	MgO	CO_2	P_2O_5	MnO	Na_2O	K_2O	H_2O^-	LOI
47.4	16.3	0.9	3.02	6.24	9.68	4.26	6.2	0.18	0.13	4.68	0.15	0.34	0.9

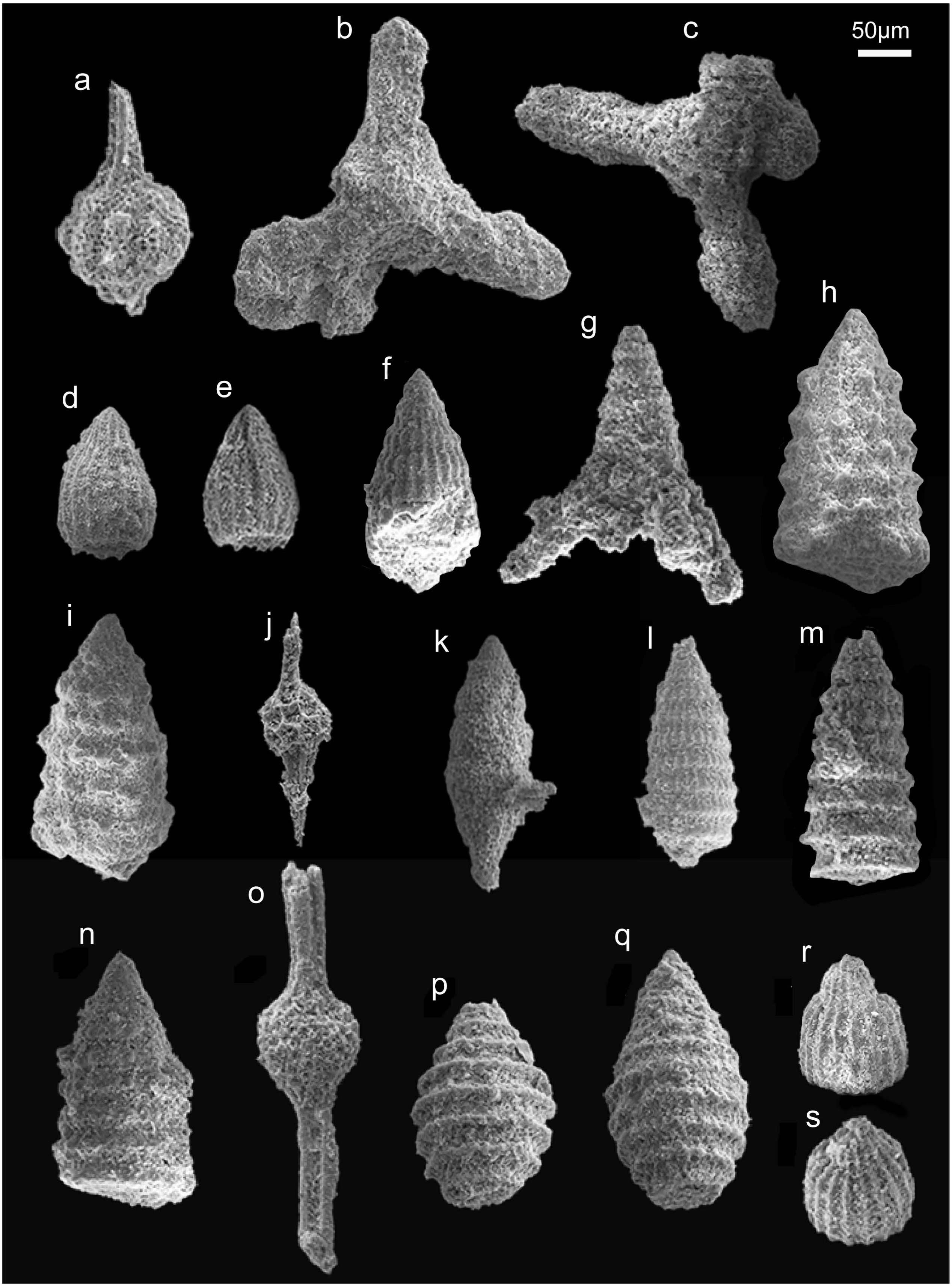

Fig. 4 - Scanning electron micrographs of radiolaria yielded from sample St11.13. a) *Acaeniotyle umbilicata* (Rüst); b-c) *Angulobracchia portmanni* Baumgartner; d-e) *Archaeodictyomitra mitra* Dumitrica; f) *Archaeodictyomitra pseudoscalaris* (Tan) *sensu* Baumgartner et al. (1995a); g) *Crolanium bipodium* (Parona) *sensu* Dumitrica et al. (1997); h) *Novixitus robustus* (Wu) *sensu* Dumitrica et al. (2022); i) *Novixitus* sp. cf. *N. robustus* (Wu) *sensu* Dumitrica et al. (2022); j) *Pantanellium squinaboli* (Tan); k) *Pseudoeucyrtis* sp. cf. *P. tenuis* (Rüst) *sensu* Dumitrica et al. (1997); l) *Pseudodictyomitra suyarii* Dumitrica; m) *Pseudodictyomitra* sp.; n) *Pseudodictyomitra* sp. cf. *P. lilyae* (Tan); o) *Stylosphaera* (?) *macroxiphus* (Rüst) *sensu* Baumgartner et al. (1995a); p) *Tethysetta ovoidala* Dumitrica; q) *Tethysetta* sp. cf. *T. boesii* (Parona); r-s) *Thanarla* sp. cf. *T. pacifica* (Nakaseko and Nishimura); same specimen, r) view from the side, s) view from the top.

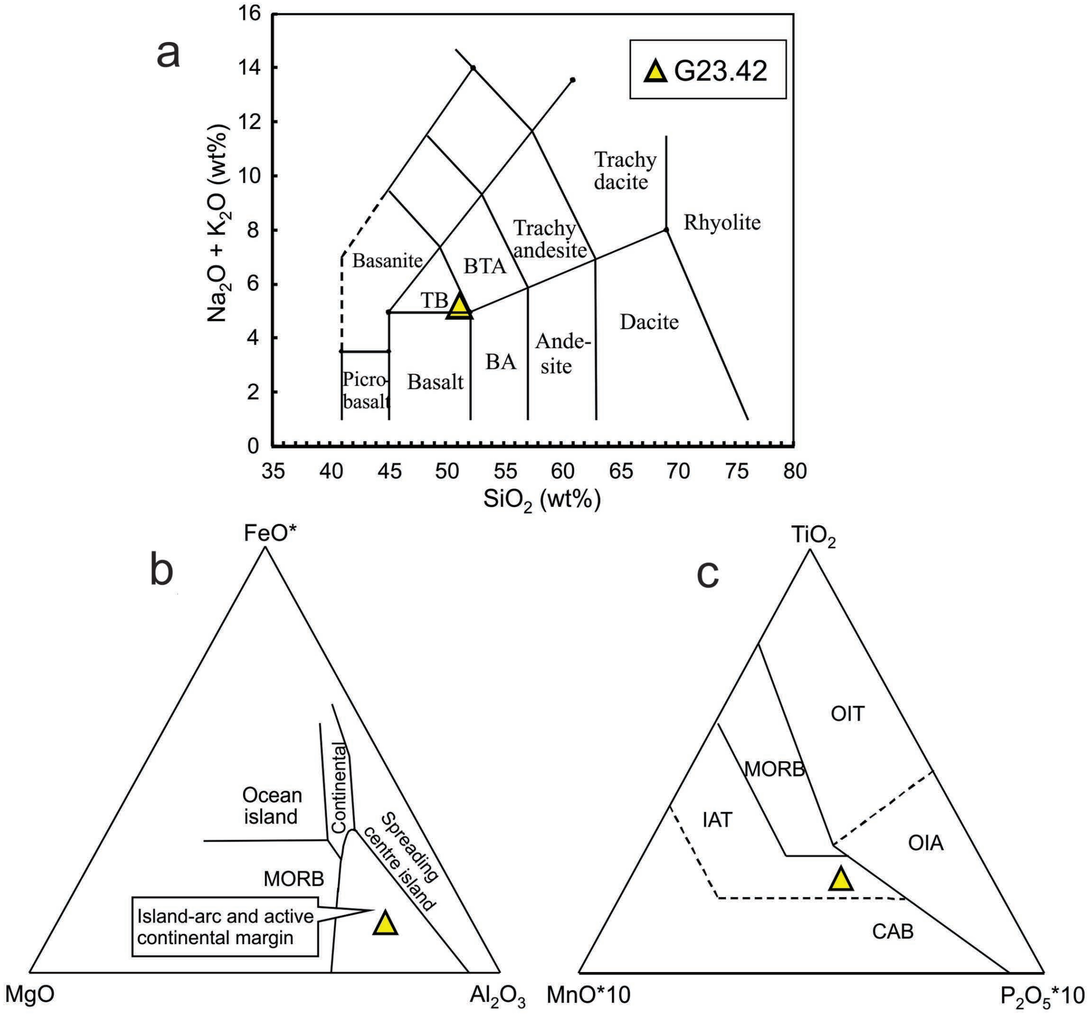

Fig. 5 - Geochemical plots of major element analyses made on sample G23.42. a) TAS diagram; b-c) major oxide-based tectono-magmatic discrimination diagrams for basalts according to Pearce et al. (1977) and Mullen (1983), respectively.

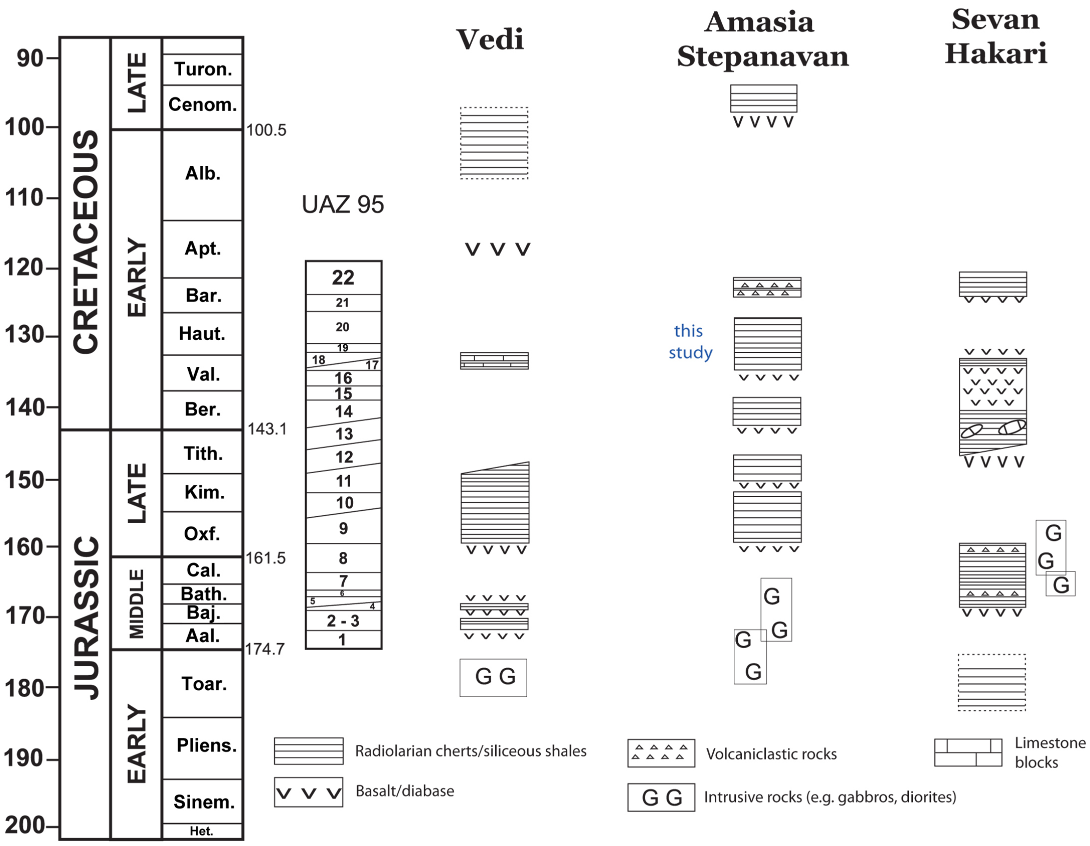

Fig. 6 - Synthesis of all known ages (both biochronological and geochronological) in the ophiolitic units of Armenia, including the results of this study from the Bazoum region (modified after Danelian et al., 2023). Time scale according to Gradstein et al. (2020).

CONCLUSIONS

The age of the radiolarites dated in the present study may be restricted to the late Valanginian - Hauterivian based on the co-occurrence of the species *Archaeodictyomitra pseudoscalaris* (Tan), *Crolanium bipodium* (Parona) and *Pseudodictyomitra suyarii* Dumitrica, which are first known to occur since the late Valanginian and of *Tethysetta ovoidala* Dumitrica, which is last known to occur in the Hauterivian. This new age evidence provides a more coherent picture for the volcanic and sedimentary evolution of the Tethyan oceanic realm preserved in the Amasia - Stepanavan sector of the Lesser Caucasus ophiolites. Taking into account the subduction-related island arc signature of the aphyric mafic lavas underlying the dated radiolarites and the presence of upper Barremian marine pyroclastics near Amasia, we propose that during the Valanginian to Barremian time interval the Amasia-Stepanavan oceanic realm was under the influence of a subduction-related island arc geodynamic regime.

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