



Geological Mapping, Stratigraphy, Palaeontology and Mineralizations of the Palaeozoic Around Valério's Quarry and the Museum of Trilobites (Arouca Geopark), Canelas, Northern Portugal

Manuel Figueiredo¹⁾, Helena Couto^{2*)}, Manuel Valério^{3)†}

¹⁾ Museum of Trilobites, Centre of Geological Interpretation of Canelas, Canelas de Cima Cx 213, 4540-252 Arouca, Portugal; email: manueffigueiredo@live.com.pt; <https://orcid.org/0009-0009-1860-4519>

^{2*)} University of Porto, Faculty of Sciences, Department of Geosciences, Environment and Spatial Planning, ICT-Institute of Earth Sciences, Rua do Campo Alegre 687, 4169-007 Porto, Portugal; email: hcouto@fc.up.pt; <https://orcid.org/0000-0002-0079-8759>

³⁾ Museum of Trilobites, Centre of Geological Interpretation of Canelas, Canelas de Cima Cx 213, 4540-252 Arouca, Portugal

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Abstract

The studied area, a sector in the reverse limb of the Valongo Anticline adjacent to the Museum of Trilobites and to Valério's Quarry (Canelas, Arouca), is located in the Central-Iberian Zone of the Iberian Massif. The main objective of this study was to obtain a detailed geological map, through the recognition of the lithologies, stratigraphic units, fossils, structures and mineralizations. The Palaeozoic succession from Cambrian to Carboniferous was studied, with the exception of the Devonian, which does not outcrop in the area. Regarding lithologies, a succession of volcano sedimentary layers has been found in Cambrian (Montalto Formation, Middle Cambrian) and in the Lower Ordovician (Santa Justa Formation), either underlying Floian massive quartzites (Tremadocian?) or overlying Floian massive quartzites (Floian). At the top of the Middle Ordovician (Valongo Formation, Dapingian-Darriwilian), near the contact with the Upper Ordovician (Sobrido Formation, Hirnantian), a ferruginous layer with abundant siliceous nodules occurs. In the base of the Sobrido Formation, massive quartzites underlie diamictites. Concerning the fossil record, abundant icnofossils such as *Cruziana* and *Planolites* were identified in the quartzites of the Lower Ordovician. Also, different somatofossils occur in the Middle Ordovician slates, namely trilobites (between the largest trilobites in the world) and other arthropods, graptolites, cephalopods, brachiopods, gastropods, bivalves and echinoderms (cystoids and crinoids). In Silurian, besides quartzites (Rhuddanian), light grey slates bearing graptolites, namely *Monograptus* (*Llandovery*), are dominant. In Carboniferous (Gzhelian) breccias, sandstones and slates with plant fossils, namely with articulated plants and ferns, occur. Regarding the mineral resources of the area, beyond the slates of Middle Ordovician (Valongo Formation) exploited in Valério's Quarry, there is a Roman mining work, the Gralheira d'Água mine (Au-As type mineralizations), associated to the Lower Ordovician (Santa Justa Formation) quartzites and volcano sedimentary layers, exploited for gold. Antimony mineralizations (Sb-Au type mineralizations) associated to the volcano-sedimentary succession of the Cambrian-Ordovician transition, were found and studied by Scanning Electron Microscopy (SEM). The final work resulted in a geological map, covering an area of 1200m by 900m, at a scale of 1: 3,400.

Keywords: geological map, palaeozoic, mineralizations, Valério's Quarry, Museum of Trilobites, Canelas

Introduction

This study focuses on the geological mapping of the area around Valério's Quarry and the Museum of Trilobites (Canelas, Arouca), a segment of the reverse limb of the Valongo Anticline, located in the Central-Iberian Zone of the Iberian Massif [1]. Being in the neighbourhood of the Museum of Trilobites and of the Centre of Geological Interpretation of Canelas, well known for its fossil exhibition as an in-situ museum, the need of a more detailed geological map to help the interpretation of future studies has become relevant. So, the main objective was to elaborate a detailed geological map, while identifying lithologies, stratigraphic succession, structures, mineralizations and fossils. Detailed field work was developed. Comparison was made with the stratigraphy, palaeontology and mineralization data published in previous studies in Valongo Anticline. Volcanic layers were identified. Scanning electron microscope study allowed the recognition of two types of mineralization.

The area is covered by the Geological map of Portugal in the scale of 1/50,000, sheet 13-B (Castelo de Paiva), 1963 [2].

Geological and Structural Setting

The Valongo Anticline runs NW-SE, dips 5° to 15° NW extending approximately 90km between Esposende and Castro Daire. This structure, formed during the first phase of the Variscan orogeny is an asymmetrical antiformal anticline. The eastern limb dips around 35° NE and runs for 20km from Valongo to Castelo de Paiva, where it is intruded by variscan granites. The western reverse limb is sub vertical and extends for 50km from Valongo to south Castro Daire where it is also intruded by variscan granite [3].

Couto [4], recognized two main episodes of folding in the area, one ante-Gzhelian and the other post-Gzhelian. The Palaeozoic succession range from the Cambrian to the Carboniferous (Fig. 1).

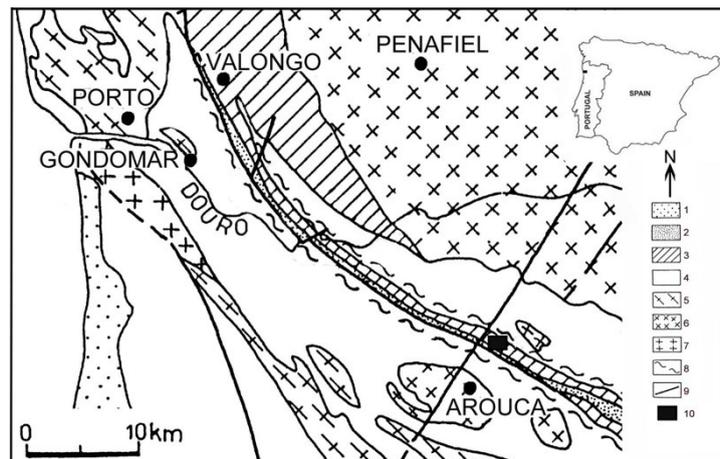


Fig. 1. Geological and structural setting of the study area. 1- Quaternary; 2- Carboniferous; 3- Ordovician to Devonian; 4- Cambrian; 5- syn- to late- F3 granitoids; 6- late- to post- F3 synorogenic biotite granitoids; 7 - post- Gzhelian granite; 8- shear-zone; 9 – fault; 10- studied area (modified after Couto & Roger [5])

In the Cambrian, interbedded slates, wackes, quartzites, quartz conglomerate and volcanic rocks occur [4], [6], [5]. Montalto Formation [4], is represented in the studied area. Interbedded volcano-sedimentary layers occur, in an association with quartzites, slates and wackes. Volcanic rocks exhibit a bimodal composition. Acid volcanism consists of intercalations of volcanoclastic rocks of rhyolitic affinities. Basic volcanism is represented by black and green volcanic rocks, weathered, in thick layers [6]. Zircon (U-Pb) secondary ion mass spectrometry (SIMS) allowed to date rhyolite occurring in the top of Montalto Formation of Middle Cambrian [7].

In Ordovician, three Formations were distinguished [8], [9]. Couto [6], considered an Early Ordovician volcano-sedimentary succession with interbedded conglomerates, of likely Tremadocian age, underlying the Lower Ordovician massive quartzite. The Santa Justa Formation of Tremadocian?-Floian age (Lower Ordovician) unconformably overlying the Montalto Formation, presents basal conglomerates and massive quartzite with trace fossils, followed by volcano-sedimentary interbedded layers. The quartzites show organic structures like Cruziana, Vexillum, Skolithos and Planolites [10] and sedimentary structures like ripple marks. Black ferruginous layers are interbedded in a succession of quartzites, wackes and slates [4]. In the Valongo Formation of Dapingian-Darriwilian (Middle Ordovician) slates prevail. To the top of Valongo Formation several siliceous nodules occur and a ferruginous horizon evidences a hiatus (paraconformity), missing upper Darriwilian, Sandbian and Katian strata (upper Middle Ordovician-lower Late Ordovician) from the succession [4], [9]. This Formation is characterized for its great paleobiodiversity, namely for the presence of trilobites and other arthropods, graptolites, brachiopods, mollusks (nautiloid cephalopod, hiolitid, rostroconchia), machaeridians (Plumulites), cnidaria, gastropods, bivalves and echinoderms (cystoids, crinoids and ophiuroids) [11], [12]. Kimmig et al. [13] described the first Middle Ordovician (Dapingian- Darriwilian) soft-bodied fossils (discoidal fossil uncertainly Patanacta, wiwaxiid sclerites, and a likely pseudoarctolepid arthropod) from northern Gondwana, found in S. Pedro da Cova and Belói (Gondomar). Neto de Carvalho et al. [14] reviewed microbial-related biogenic structures from the slates of Valongo Formation in Canelas, with the description of the ichnoassemblages. The Sobrido Formation of Hirnantian age (Upper Ordovician) is represented by two members [8], [9]. The lower one is mainly composed of quartzites, which overlay the slates with nodules from the Valongo Formation. The transition to the upper member evidences an erosive contact marked by a ferruginous horizon and followed by thin laminated siltstones. The top of this unit is variable in thickness and contains both laminated and massive diamictites, interbedded with quartzites, conglomerates and slates evidencing dynamic changes in sedimentary and geochemical processes in response to palaeogeographic changes during the Upper Ordovician and transitional to the Lower Silurian on the north Gondwana platform [9]. Silurian black quartzites (Rhuddanian) and light grey slates bearing graptolites (Llandovery) occur overlying the diamictites [9]. The Lower Devonian is formed by fossiliferous pale grey slates interbedded with thin quartzites [15]

The Carboniferous is represented by breccias-conglomerates, sandstones and fossiliferous slates from the Lower Gzhelian [16].

In the studied area gold and antimony mineralizations occurs, being part of the Dúrico-Beirão Mining District [4].

Fieldwork- Geological Mapping, Stratigraphy and Palaeontology

The Palaeozoic succession from Cambrian to Carboniferous was analysed, with the exception of the Devonian, which does not outcrop in the area [Fig. 2 (a)]. The elaboration of the geological map required the realization of fieldwork. In the area it was possible to identify geological contacts, lithostratigraphic units, structures and to characterize the stratigraphic succession. About half a hundred GPS points were acquired during the fieldwork allowing proposing a detailed geological map.

Cambrian is represented by the Montalto Formation [4]. In the western part of the map, western of the reverse limb of the Valongo Anticline, Cambrian yellowish slates with interbedded quartzites are in contact with the Carboniferous. In the core of the Anticline, in the eastern part of the map, Montalto Formation underlie the Lower Ordovician massive quartzites. In the transition Cambrian to Ordovician we can observe interbedded quartzites, slates and volcanic rocks. Green volcanic rocks with interbedded siliceous layers showing mineralization were identified [Fig. 2 (b)].



Fig. 2. (a) Palaeozoic succession from Cambrian to Carboniferous around Valério's Quarry (photo from Lower Ordovician quartzitic ridge to the Upper Ordovician quartzitic ridge) (b) Green volcanic rocks with interbedded siliceous layers of likely Tremadocian age underlying massive Floian quartzites

In the Lower Ordovician (Santa Justa Formation) metric light quartzites interbedded with dark millimetric pelitic layers originate the main ridge of the area [Fig. 3(a)]. Different trace fossils, namely *Cruziana* developed in the stratification plane (S0) and numerous tubular trace fossils identified as *Planolites* [Fig. 3(b)]. Sedimentary structures are also abundant and current ripples with cross lamination and load casts were observed [Fig. 3c]. To the top of Santa Justa Formation interbedded volcano sedimentary layers of Floian age occur [Fig. 3(d)].

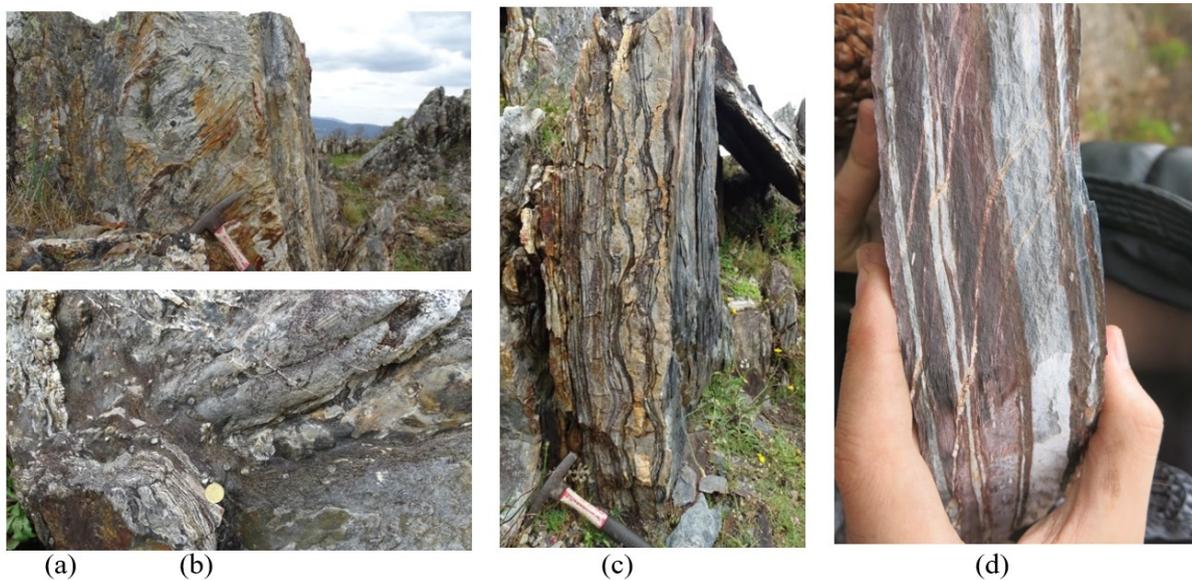


Fig. 3. (a) Massive Floian quartzites exhibiting cross stratification; (b) *Cruziana* and *Planolites* in Floian quartzites; (c) Interbedded Floian quartzites and slates exhibiting current ripples with cross lamination and load casts. (d) Interbedded volcano sedimentary layers of Floian age

In Santa Justa Formation, Roman mining works occur. Gralheira d'Água Mine resulted from the exploitation of auriferous quartz veins, installed in the Floian massive quartzite.

In the Middle Ordovician (Valongo Formation, Dapingian/Darriwilian), ardosiferous slates are dominant. At the top of the succession numerous siliceous and phosphate nodules occur followed by a ferruginous layer [Fig.4(a) and (b)].



Fig. 4. (a) Slaty of Valongo Formation with joints and oxidation (b) Slaty with ferruginous horizon and nodules evidencing an erosional contact between the Valongo Formation and the Sobrido Formation

Among the very abundant and varied invertebrate fossils of Valongo Formation, trilobites, graptolites, cephalopods, brachiopods, gastropods, bivalves and echinoderms (cystoids and crinoids) were identified (Fig. 5).

Trilobites are considered by the scientific community to be among the largest in the world. Samples bearing several trilobites in monospecific concentrations suggest gregarious behavior in these arthropods.

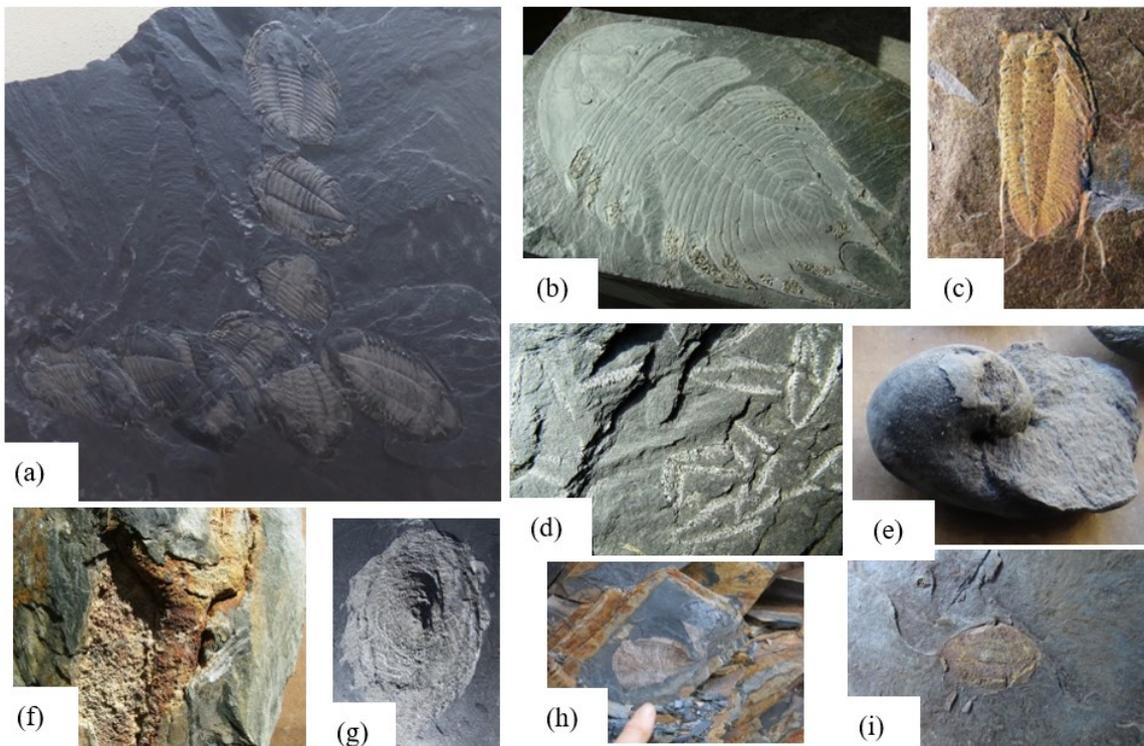


Fig. 5. Fossils of Valongo Formation, Middle Ordovician, Museum of Trilobites, (a) monospecific concentrations of trilobites *Asaphellus toledanus*, internal moulds (l=13cm); (b) trilobite, *Hungioides bohemicus*, internal mould (l=35cm); (c) trilobite *Bathycheillus castilianus*, internal mould (l=6cm);(d) graptolite, *Didymograptus artus*, external mould (l=2cm);(e) gastropod *Sinuities*, internal mould (l= 3 cm); (f) cephalopod nautiloid, internal mould showing septa and siphonal canal (l= 17 cm); (g) echinoderm, cystoid, internal mould (l=3,5cm); (h) brachiopod, external mould (l=4,5cm) (i) bivalve *Babinka*, internal mould (l=4cm) (l=longest length)

In the Upper Ordovician (Sobrido Formation, Hirnantian) basal massive quartzites occur [Fig. 6 (a)]. These are rejected by faults, joints and have a lenticular nature. Overlying the quartzites, massive diamictites [Fig. 6 (b)]. has ferruginous crusts showing iron diffusion textures or filling the joints. Laminated diamictites were also observed.



Fig. 6. (a) Hirnantian massive diamictites affected by joints filled with quartz (b) Dropstone in diamictite

In this area, Silurian is dominantly represented by laminated micaceous light grey slates, interbedded with centimetric to metric quartzitic layers [Fig. 7(a)]. Slates are very soft and easy to cleave by hand [Fig. 7(b) and (c)].

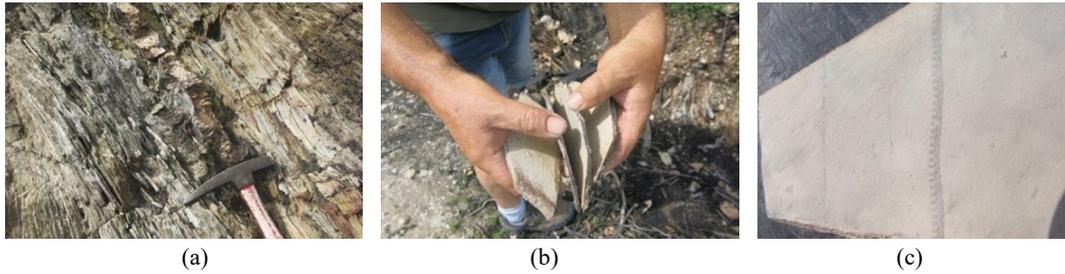


Fig. 7. (a) Silurian laminated light grey slates with interbedded centimetric quartzitic layers and a vein of quartz in the stratification plane. (b) and (c) Laminated Silurian light grey slates with Monograptus.

The graptolites present in these slates indicate a Llandovery age. Centimetric to metric quartzitic layers occur interbedded with light grey slates.

In the mapped area the Carboniferous of continental facies lies between the Silurian and the Upper Ordovician and between the Silurian and the Cambrian. A control by faults is evidenced by the brecciation of the rocks and by the abundance of iron oxides. Breccias and conglomerates prevail with interbedded sandstones and slates [Fig. 8(a) and (b)]. Carboniferous basal breccia conglomerate is usually clast-supported with pelitic matrix and clasts of different lithologies, namely of quartzites, lydites, slates and quartz, mostly angular, with dimensions varying from coarse sand to boulders. Sedimentary structures as channel-filling of braided rivers were observed. Slates contain fossils of plants, namely articulated plants and ferns.



Fig. 8. (a) Carboniferous basal breccia conglomerate; (b) Carboniferous interbedded sandstones and slates

Mineralizations in the Studied Area

Regarding mineral resources, other than the slates of Middle Ordovician exploited in Valerio's quarry, there is an ancient Roman mining work, the Gralheira d'Água mine, associated with Floian quartzites, with interbedded slates and volcanics of the Lower Ordovician that was exploited for gold [17]. An auriferous quartz vein, N160 trending, cutting the Floian massive quartzite, was exploited.

Gralheira d'Água mine belongs to the Dúrico Beirão Mining District. Couto [4] distinguished four paragenetic associations: Sn-W, Sb-Au, Au-As and Pb-Zn (Ag). Gralheira matches the Au-As type, in which gold appears associated with arsenopyrite and pyrite, being the preferentially exploited mineralizations by the Romans (Fig. 9).



Fig. 9. Gralheira d'Água mine, showing narrow cavity, corresponding to the extracting of an auriferous quartz vein in the Floian massive quartzites with interbedded volcano-sedimentary layers

As said before, in the vicinity of the Gralheira d'Água mine, mineralization occurs next to the Cambrian-Ordovician contact, in green volcanic rocks with interbedded siliceous layers. The small metallic minerals were observed under a binocular magnifying glass and under a Scanning Electron Microscopy (SEM). In a first observation pyrite and stibnite were identified.

Scanning electron microscopy (SEM) study

The electron microscope analysis was performed using a high-resolution Scanning Electron Microscope, with X-Ray Microanalysis: JEOL JSM 6301F/ Oxford INCA Energy 350, at the Centro de Materiais da Universidade do Porto (CEMUP) (analyst Dr Daniela Silva). The samples were coated with a thin film of C, by vaporization, using the JEOL JEE – 4X Vacuum Evaporator equipment.

Three samples of mineralized quartz veins associated to green volcanic rocks (Tremadocian?) were selected for study (Samples 587 A, B and C).

In the SEM, smaller grains of metallic minerals of yellowish color (sample P. 587A) were identified as pyrite. The presence of iron oxides was also noticed. In the same sample, a Pb, Fe and Al phosphate was identified as plumbogummite ($\text{PbAl}_3(\text{PO}_4)(\text{PO}_3\text{OH})_3(\text{OH})_6$) (Fig. 10).

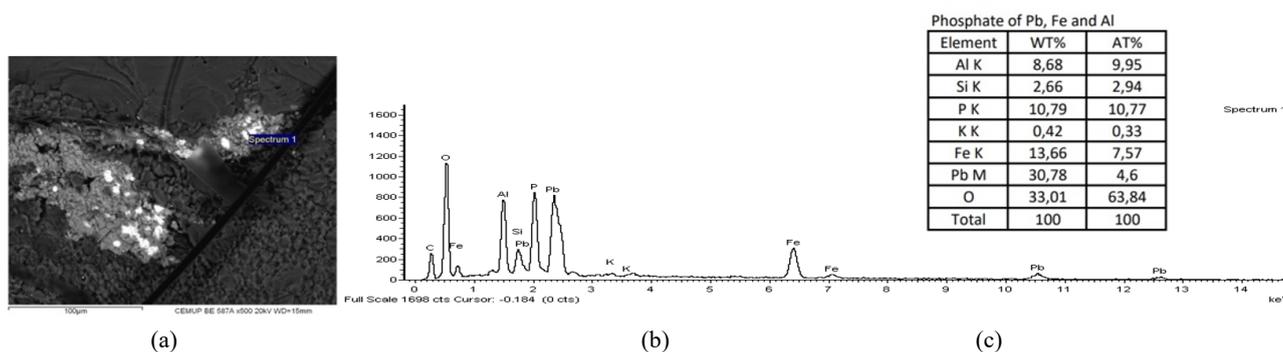


Fig. 10. Scanning electron microscopy (SEM) images (a), spectrum energy dispersive spectroscopy (EDS)(b) and semi-quantitative analysis (c) in a mineralized quartz vein. The analyzed mineral is a phosphate identified as plumbogummite ($\text{PbAl}_3(\text{PO}_4)(\text{PO}_3\text{OH})_3(\text{OH})_6$) (sample P. 587A)

In the samples P. 587B and 587C, SEM images and spectra, showed the destabilization process of berthierite in stibnite, corresponding the lighter colors to stibnite (Sb_2S_3) (spectrum 2) and darker colors to berthierite (FeSb_2S_4) (spectrum 3) with variable content in Fe (Fig. 11). It is thus stibnite II resulting from the alteration of berthierite by loss of Fe. The presence of Sb – O+Sb oxides was also noted (Figure 11).

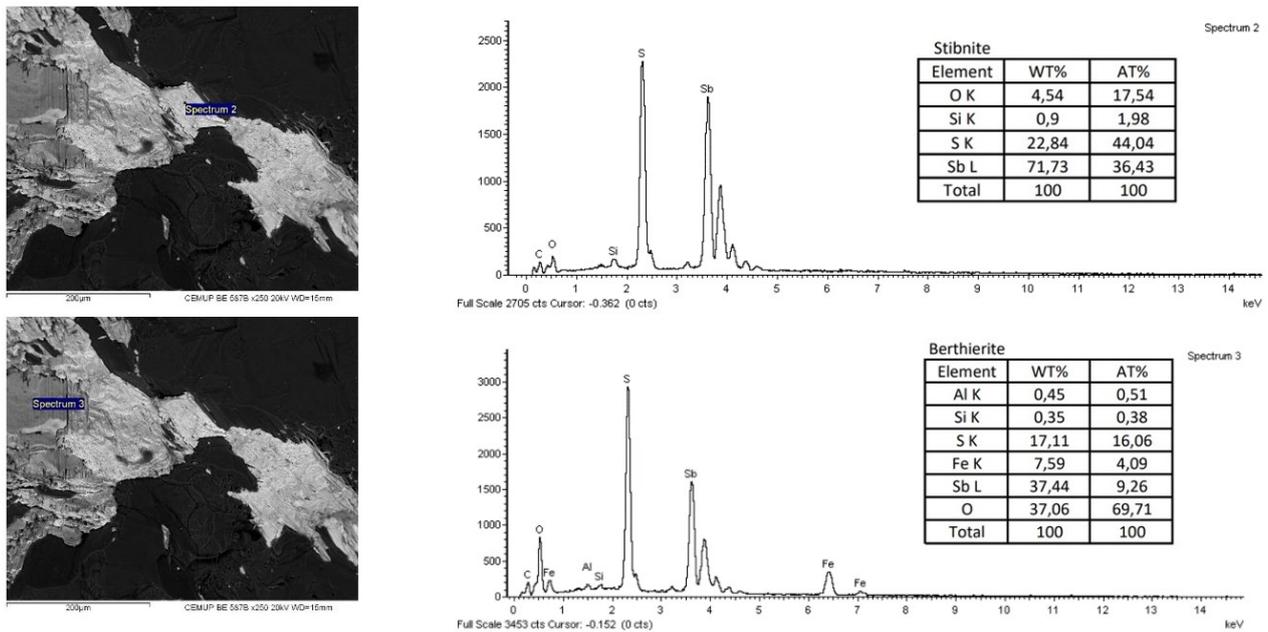


Fig. 11. Scanning electron microscopy (SEM) images (a), spectrum energy dispersive spectroscopy (EDS) (b) and semi-quantitative analysis (c) in a mineralized quartz vein

Results and Discussion

Fieldwork, based on the lithologies, structures, stratigraphic and palaeontological data and on the petrographic studies complemented with the SEM, allowed to perform a detailed geological map at a scale of 1: 3,400 (Fig. 12).

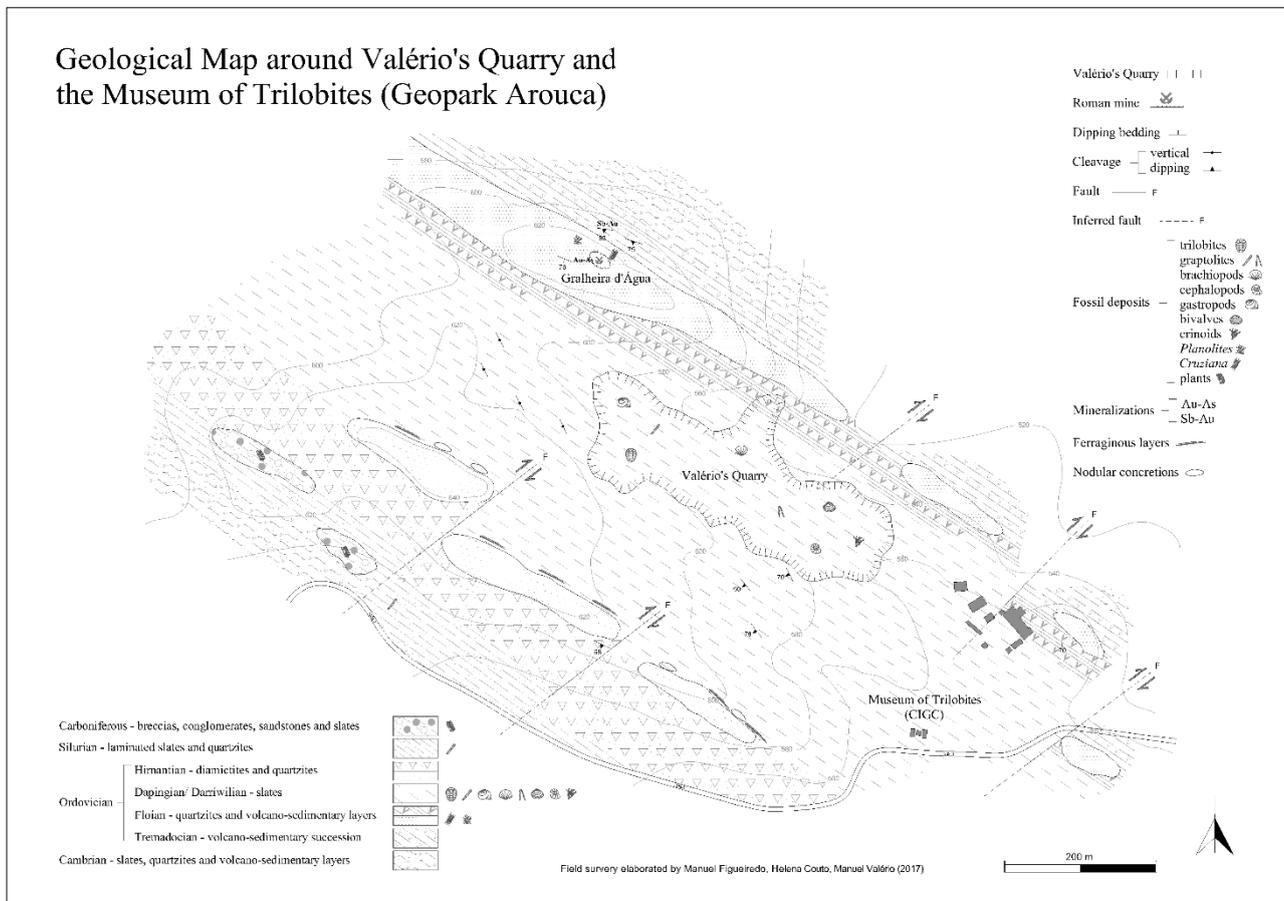


Fig. 12. Geological map of the Palaeozoic around Valério's quarry and the Museum of Trilobites

In the transition Cambrian to Ordovician interbedded quartzites, slates and volcanic rocks namely the green volcanic rocks are equivalent to the succession Cambrian–Lower Ordovician described by Couto [6] and Couto & Roger [5] in the Valongo Anticline.

Abundant trace fossils such as *Cruziana* and *Planolites* occur in the Lower Ordovician (Floian) quartzites. These layers exhibit disturbed stratification related to deposition in very unstable conditions, with bioturbation, ripple marks and load structures.

Different studies concerning the invertebrate fossils of the Canelas slates confirm the exceptionality and diversity of this fossil fauna [18], [12], [19], [20], [21], [22]. Trilobites, graptolites, brachiopods, mollusks (nautiloid cephalopod, hiolitid, rostroconchia), cnidaria, ostracods, gastropods, bivalves and echinoderms (cystoids, crinoids and ophiuroids) occur. Trilobites, with sixteen genera identified till now, are considered by the scientific community to be among the largest in the world.

When comparing the succession of Valongo Formation in the studied area with the same Formation outcropping more to north in Valongo and Gondomar, described by Couto [4], [12], it seems that the Valongo Formation in Canelas is more incomplete lacking tournemini and borni biozones, being *Placoparia cambriensis* the only species of *Placoparia* described till now in Canelas [19], [22].

At the top of the Valongo Formation, a ferruginous layer and numerous siliceous and phosphate nodules occur in contact with the Sobrido Fm. According to Couto et al. 2013 these ferruginous layers evidence a hiatus (paraconformity), missing upper Darriwilian, Sandbian and Katian strata (upper Middle Ordovician–lower Late Ordovician) from the succession. After Loi & Dabard [23] the genesis of these nodules is related to the fluctuations in terrigenous flux due to eustatic variations. During periods of sea level rise sedimentation is dominated by bioclastic elements (calcareous, siliceous and phosphatic), decreasing the terrigenous clastic supply.

Silurian slates, with interbedded cherty or quartzitic layers bearing graptolites are comparable with those of Middle to Upper Llandovery (Lower Silurian) age described by Romariz [24].

Carboniferous breccia-conglomerate, sandstones and slates observed in small outcrops west of the reverse limb of Valongo Anticline, show a succession similar to the one observed more to north [16], [25], [4].

Regarding gold-antimony mineralizations of the Dúrico-Beirão mining district, Couto [4] distinguished two types: Sb- Au and Au-As besides two gold impoverished types that are also present: Pb-Zn-(Ag) and W-Sn. The study by SEM allowed to corroborate that, in addition to the Au-As mineralizations previously reported at Gralheira d'Água Mine [17] where gold occurs associated to the sulfides, namely arsenopyrite and pyrite, Sb-Au mineralizations in which gold appears associated with stibnite are also present. The study of the antimony mineralizations now identified in the studied area shows their association with the volcanic layers of the transition Cambrian – Lower Ordovician as proposed by Couto et al. [26]. for this type of mineralization. Scanning electron microscopy (SEM) also allowed to identify for the first time the presence of plumbogummite ($\text{PbAl}_3(\text{PO}_4)(\text{PO}_3\text{OH})_3(\text{OH})_6$) a Pb, Fe and Al phosphate in the Dúrico-Beirão mining district.

Conclusion

The works developed in the field complemented with laboratorial research, resulted in the elaboration of a detailed geological map, covering an area of 1200m by 900m around Valério's Quarry and the Museum of Trilobites.

This study may be a contribution to future studies, in particular to the palaeontological studies that are going on, in this Museum. Regarding lithologies and stratigraphy, present study allowed the recognition of volcanic rocks, not described in the sheet 13-B (Castelo de Paiva) of Geological map of Portugal in the scale of 1/50,000, though identified in other parts of Valongo Anticline, namely in Fragas da Torre (Arouca) [5].

A new phosphate of Pb, the plumbogummite ($\text{PbAl}_3(\text{PO}_4)(\text{PO}_3\text{OH})_3(\text{OH})_6$) was recognised being a contribution to the mineral species list of the mineral deposits studied in Dúrico-Beirão mining district.

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