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## Later Prehistoric tin mining in the Ervedosa Mine (Vinhais, Portugal): evidence and context

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### Abstract

This paper presents a comprehensive study of the evidence for ancient tin mining at the Ervedosa mine (Vinhais, Portugal). The geological context of the site indicates a rich cassiterite (SnO<sub>2</sub>) deposit, which was subject to mining in the 20th century. Some ancient mining and ore processing stone tools were recovered during the 20th century mining operations, namely one hammer, one pounder, one flat anvil and five small tools used both as pounders and crushing anvils, evidencing Prehistoric mining activities. XRF and SEM-EDS chemical analyses were performed on primary and secondary cassiterite samples from the mining site, demonstrating the abundance and chemical heterogeneity of the tin (Sn) ores. The stone tools can be ascribed to Bronze Age or, at the latest, Early Iron Age (2nd millennium to first half of 1st millennium BCE) by comparison with similar tools from other Iberian and European archaeological contexts. High-resolution

photogrammetric 3D models of the tools are made available in this study. The historical descriptions of the findings and the research made on the technical archives about the mine have allowed correlating the tools to mining in a primary context, focused on rich quartz veins in granitic or greisen bedrock, rather than mining in a secondary context. XRF and SEM-EDS analyses performed on the stone material and on surface adherences support their identification as specific types of hard rocks, such as granite, amphibolite and quartzite. They also allowed the detection of Sn-rich adherent particles, confirming their use for Sn-material processing. The potential relation between the cassiterite resources and the local Later Prehistoric (Bronze Age to Early Iron Age) settlement pattern is also discussed. The results raise awareness and provide relevant data about the existence of tin mining in primary contexts during Later Prehistoric times in the NW Iberian Peninsula.

### **Keywords**

Cassiterite, Tin mining, Iberian Peninsula, Bronze Age, Stone tools, Ore processing.

### **1 Introduction**

The Ervedosa tin (Sn) mine, also known as Tuela mine, is located in Vinhais, in the Trás-os-Montes region, NE Portugal (Fig. 1). Its recent history is well documented thanks to rich archive collections from the Arquivo Histórico do Norte de Portugal (Direção-Geral de Energia e Geologia), from the Direção Geral do Norte (Ministério da Economia) and from the Direção de Serviços de Indústria e dos Recursos Geológicos (Fernandes 2010). These archives describe technical and economic aspects of 60 years of mining operations, from 1909 to 1969. Other official documents also contain the discovery and registration of the ore deposit by different explorers, with the earliest date of registration being in 1873 (Fernandes 2010). Since this date, the ore deposit was registered 18 more times by different explorers. In the records, most explorers stated to have found the deposit through field surveys, noting the presence of cassiterite ( $\text{SnO}_2$ ) and traces of ancient mining works.

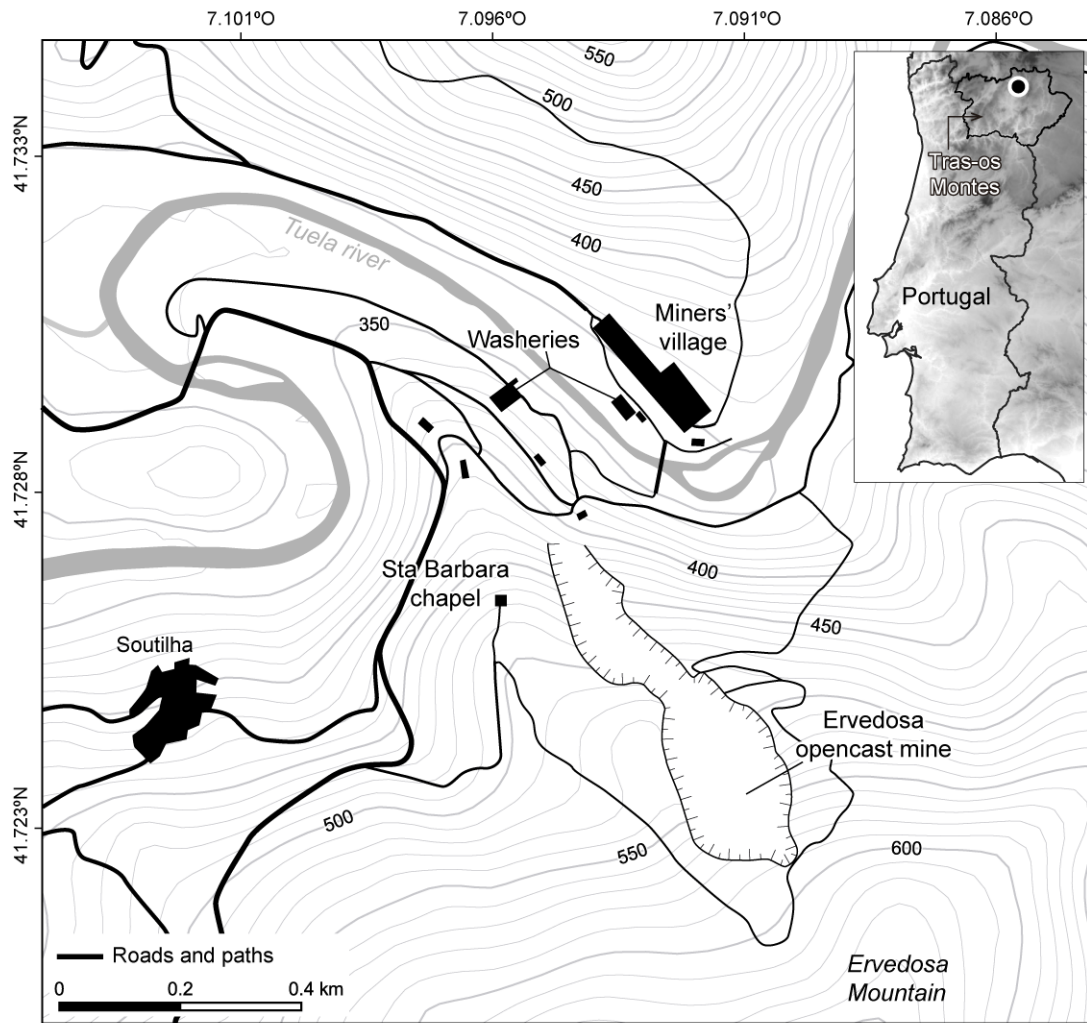


Fig. 1. Location of the Ervedosa mine in the Portuguese territory (top right) and of the opencast mine area and associated buildings related to the 20<sup>th</sup> century mining operations (close to Soutilha village).

The first industrial mining works started with the French company Société des Mines d'Étain de Ervedosa (1906-1920), followed by two English companies, the Ervedosa Tin Mines Ltd. and then the Tuela Tin Mines Ltd. From 1928 to 1969, the exploitation carried out by the Tuela Tin Mines Ltd. produced about 6000 tons of cassiterite and a similar weight of arsenic ( $As_2O_3$ ) (Gomes and Neiva 2002). All the tin was exported to England as concentrated ore of ca. 65% tin, e.g., 82,5% ore purity (Fernandes 2010). This recent mining activity had two consequences. On the one hand, it revealed a former phase of mining through the findings of several mining and ore processing stone tools (Ferreira and Castro 1949), which were related to the Later Prehistoric period in a broad sense, i.e., Bronze Age and Early Iron Age (2nd millennium and first half of 1st millennium BCE). On the other hand, it ended with any possibility of finding the old mining works today, since they were destroyed by the decision of the company to implement opencast exploitation since the mid-1930s (Fernandes 2010: 44-53) (Fig. 2). So far, these artefacts are among the very few reported archaeological cobble stone tools from tin mining site in Western Europe. This gives them a significant role in the investigations of ancient tin and bronze productions. Identifying the early sources of tin is an old question that is still unsolved (Muhly 1973; Charles 1975; Muhly 1985; Giumlia-Mair, Lo Schiavo 2003; Hauptstein et al. 2010; Berger et al. 2019). The data presented here contribute to assessing the tin production of the Iberian Peninsula which is the largest tin-bearing area of Western Europe (Comendador et al. 2017).

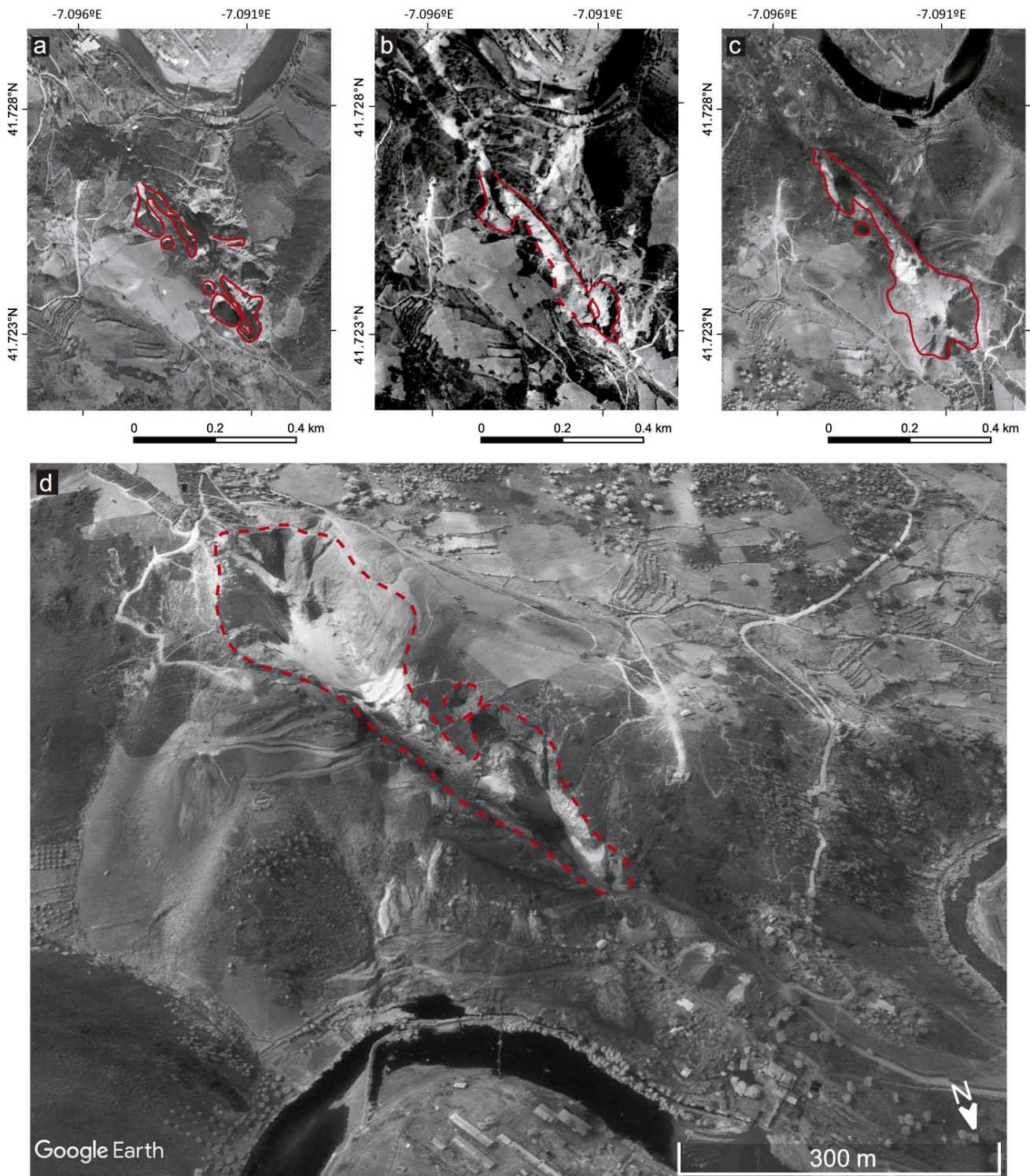


Fig. 2. Evolution of the opencast mining work in Ervedosa mine (highlighted in red in the top images) visible by the historical aerial photographs: (a) SPLAL flight 1949; (b) USAF flight 1958; (c) DGT flight 1965. At the bottom (d) oblique view of the 1965 aerial photograph projected on the Google Earth topographic base map. SPAL and USAF photographs from the Centro de Informação Geoespacial do Exército, Lisbon, Portugal and DGT photographs from the Direcção Geral do Território, Lisbon, Portugal (images were processed according to Fonte et al. 2021).

This mining site and its stone tools are important elements for understanding the type of mining resources used by the local communities during late Prehistory. The present work adopts integrated multidisciplinary research combining geological, archaeological, archival and analytical approaches, as previously defined by our team (Figueiredo et al. 2018; Figueiredo et al. 2022; Fonte et al. 2017; Fonte et al. 2021). This approach

allows to overcome the limits imposed by the destructions resulting from the recent mining and helps to shed some light on the oldest phase of the local mining activity.

In this paper, we present the chemical analyses of the stone tools and cassiterite samples, and we discuss their geological and archaeological contexts. The archaeological artefacts will bring elements to discuss the technical aspects of ancient cassiterite processing and provide a broad chronological framework for the ancient mining phase of the site. The stone tools were submitted to non-destructive *in-situ* XRF analysis to aid the identification of the stone material. Also, surface material adherent to the top of the artefacts was collected for SEM-EDS analysis to search for traces of mining or metallurgy, i.e. to detect any Sn-material that could support their use for ore processing or for slag crushing (to recover metallic prills). In addition, 3D models of most artefacts were recorded using photogrammetry, allowing easier comparisons with other ancient mining and ore processing stone tools. The cassiterite samples were subjected to *in-situ* XRF analysis, WDXRF and SEM-EDS analysis depending on the authorisation to handle the samples for research and their adequacy for the analysis, such as size and shape. The results allowed us to make a first evaluation of the cassiterite composition, and evaluate the presence of minor and trace elements. Previous research on Iberian cassiterites has highlighted the variability of minor and trace elements at regional and even local scale (Murciego et al. 1997; Neiva 2008; Figueiredo et al. 2018; Figueiredo et al. 2022), showing the need to perform analysis on several local samples to evidence a representative compositional pattern for a single site (a geochemical signature). Finally, a discussion about the mine and its relation to archaeological context will follow. At local scale (up to 10 km around Ervedosa mine), we emphasize the existence of several Later Prehistoric and Roman settlements. At the scale of Trás-os-Montes region, we notice the evidences of bronze manufacturing since Middle Bronze Age and of mining vestiges of pre-industrial chronology through all the area (Lemos 1993; Maciel 1993). At a broad scale, this study opens new perspectives about tin mining in the Trás-os-Montes region and reinforces the evidence of Later Prehistoric tin mining in Northwest Iberia.

## 2 Materials and methods

### 2.1 Cassiterite samples

Ten cassiterite samples from Ervedosa were analysed: six samples from the Museu de Jazigos Minerais Portugueses, three samples from the Geosciences Museum (IST) and one sample collected during our fieldwork, from a quartz vein at the bottom edge of the opencast mine. Regarding the geological context of sampling, seven were from quartz veins, corresponding to the primary tin sources, and three were from the alluvium of the Tuela river, which flows at the base of the mine. Archival reports show that during the 20th century, the mining work focused on the primary quartz veins, but Tuela's alluvium was not neglected and was also processed at a lower extent (Fernandes 2010; Gomes 1996). The analysed cassiterite samples are shown in Fig. 3, being evident the variability in the size (ranging from <1 cm to 10 cm) and appearance of the samples, with those of alluvial origin having more rounded surfaces.

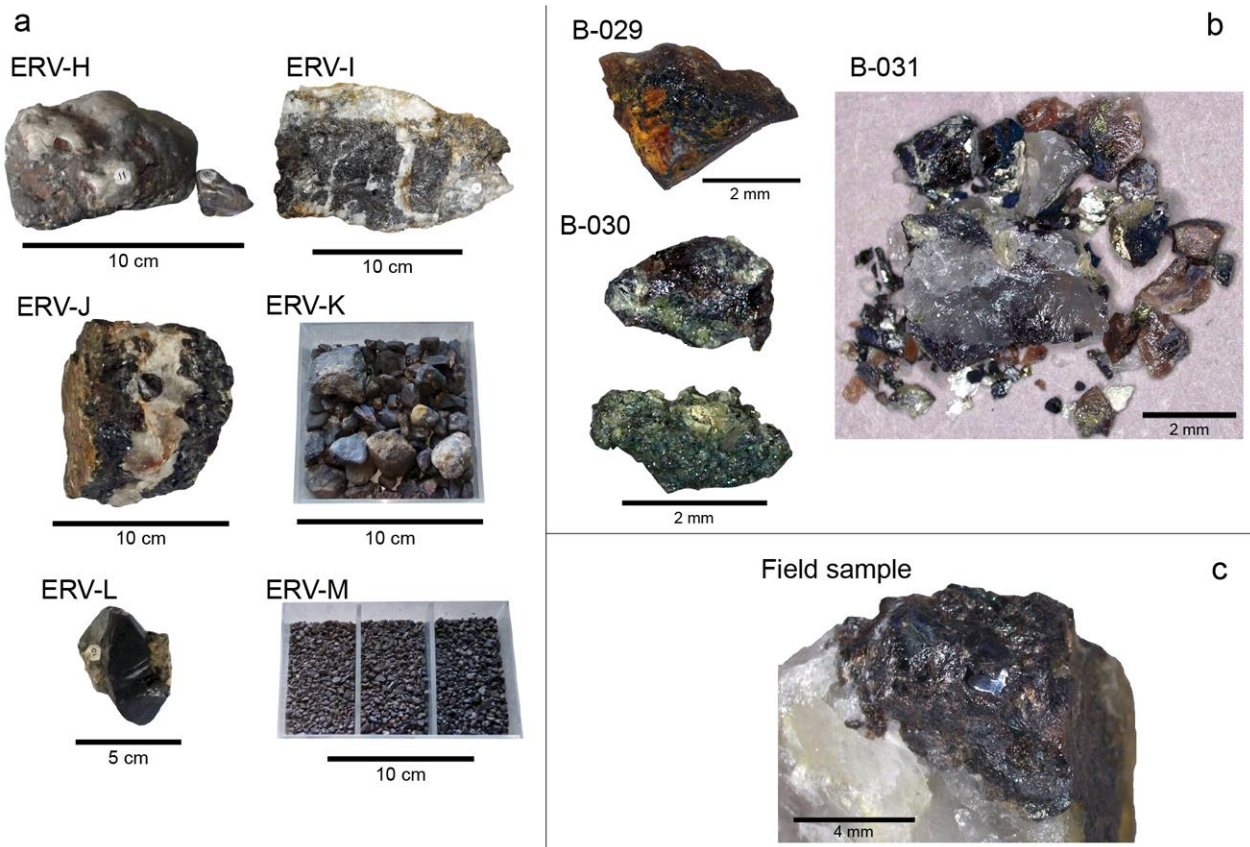


Fig 3. Analysed cassiterite ores from: (a) Museu de Jazigos Minerais Portugueses; (b) Geosciences Museum of the IST (ranging at the mm size); (c) field sample.

## 2.2 Stone ore processing and mining tools

Eight stone tools from the Ervedosa mine have been preserved (Fig. 4). Seven are deposited in the Museu de Jazigos Minerais Portugueses (Porto) and one at the Geosciences Museum of the IST (Lisbon). One of the tools from the Museu de Jazigos minerais Portugueses (ERV-C) was the first described from this site, in a 1949 publication, and considered to date to the Bronze Age (Ferreira and Castro 1949). This one and another “similar piece” is mentioned by C. Domergue after a visit to the Museum in 1965, stating that it is not possible to know if they were pre-Roman or Roman (Domergue 1987: 510). Both were considered to be of Roman date by F. Sande Lemos, who compared them with tools from the Tresminas gold mine, also in the Trás-os-Montes region (Lemos 1993, t. 1, 359-360). More recently, these tools were briefly cited in a catalogue dedicated to archaeological artefacts in the museums from the Instituto Nacional de Engenharia, Tecnologia e Inovação, with no further description (Brandão and Silva 2006: 559-560). Brandão and Silva however suggested that they could date to the Bronze Age. Lastly, six of the ore processing tools were presented through photographs with a basic description of their dimensions by C. M. B. Fernandes in her book about the recent history of the Ervedosa mine (Fernandes 2010). This author follows the interpretation of Ferreira and Castro and considers them as Bronze Age tools. During our visit to the Museu de Jazigos Minerais Portugueses, we accessed the documents containing the location of these tools in the former arrangement of the museum collections. In those documents, a stone hammer (ERV-G) was also associated with the ore processing tools from Ervedosa. We decided to include this stone hammer in the present study based on these documents and on the discussion with members of the museum. The small crushing tool preserved in the Geosciences Museum of the IST (E-01), unpublished to date, is similar to the ERV-E tool from the Museu de Jazigos Minerais Portugueses.

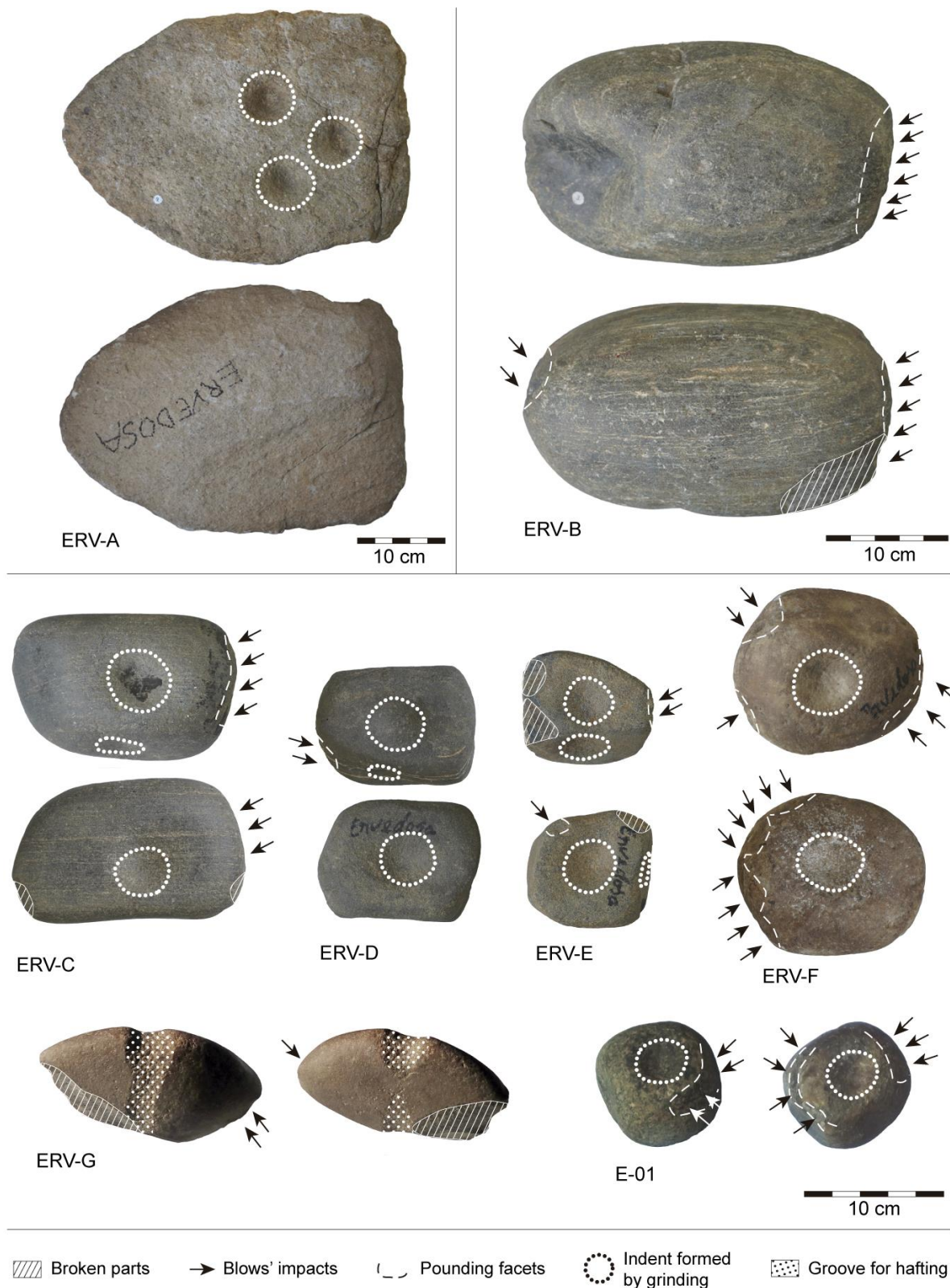


Fig. 4: Archaeological mining and ore processing stone tools from Ervedosa mine: ERV-A to ERV-G come from the Museu de Jazigos Minerais Portugueses (Porto, Portugal); E-01 from the Geosciences Museum of the IST (Lisbon, Portugal). On each artefact, areas corresponding to broken parts and to wear (as from blow impacts, pounding and grinding) are depicted.

The precise findspots of the tools are not known, except for ERV-C (Ferreira and Castro 1949): it was found above the level *Pons*, close to the gallery *Petit Delchet* (Fig. 5). Maps of the mine available in the Arquivo Histórico do Norte do Portugal from the Direção-Geral de Energia e Geologia (Amorim and Freitas 2015) show



that the location of the tool was above the 525 m level (elevation above sea level), 165 m above the Tuela river (Anonymous 1914). The discovery of this tool during the underground operations indicates the presence of previous underground galleries, which were destroyed by the progress of the 20th century mine. The finding of artefacts in ancient galleries is not uncommon. Generally, after being mined, galleries are filled naturally and/or in purpose with materials from the time of the mining and from the surface. In Portugal it is well known the case of the Late Bronze Age bronze palstave found at a depth of 12 m in a copper mine gallery in Quarta-feira (Sabugal, central Portugal) during the 19th century mining works (Melo et al., 2002). The existence of ancient galleries in Ervedosa is also described in the documents that narrate the circumstances of the discovery of the ore deposit in the 19th century, with finders referring to traces of ancient mining works. Also, more recent oral references reported by Fernandes (2010) state that small galleries, where someone could only enter on all fours, existed before 1906 on the slope where the opencast mine was dug. During our field survey, we were not able to identify any remaining old gallery, as well as no additional ancient tools. The slope with the deposit has been severely altered by the modern mining works and likely all ancient evidence destroyed.

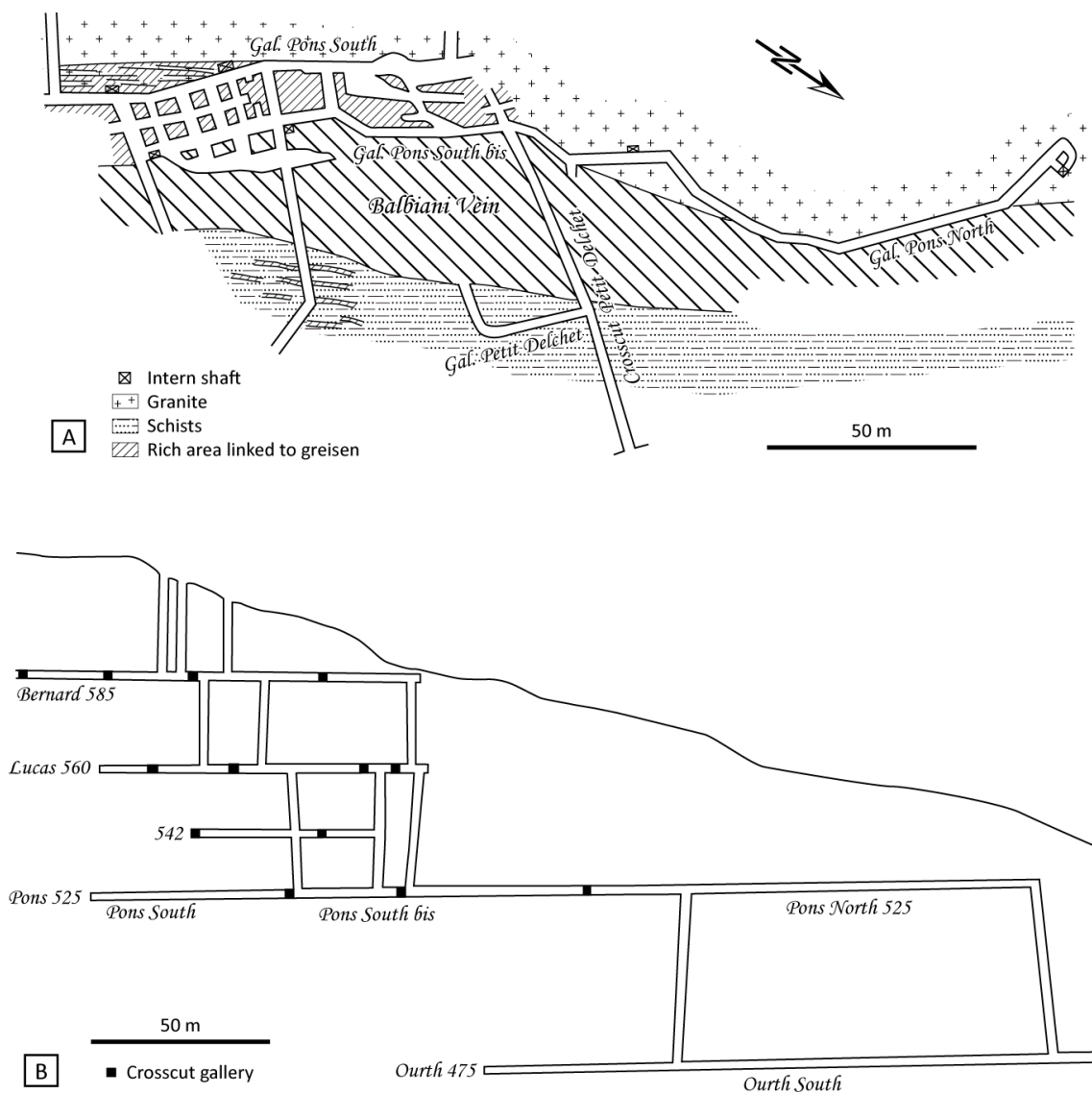


Fig. 5. Topography of the mine in 1914 with the location of the level Pons and the Petit Delchet gallery, used to indicate the location of the finding of the artefact ERV-C (adapted from Anonymous 1914): (A) plan of the level Pons; (B) longitudinal section along the Balbiani vein (the numbers refer to the elevation above sea level, in meters; the Tuela river is at 360 m). Note that the tool ERV-C was found above the level Pons, close to the gallery Petit Delchet.

### 2.3 XRF

Non-destructive X-Ray Fluorescence analyses (XRF) were performed on the stone tools and cassiterite ores in the Museu de Jazigos Minerais Portugueses (Porto, Portugal) using a portable XRF Bruker S1 Titan 600 instrument, containing a Rhodium anode tube (4 W, 15-50 KeV and 5-100  $\mu$ A) and a SSD (silicon drift detector) with resolution <145 eV. Each analysis had a duration of 90 s, and three to four analyses were made in each artefact or sample, being considered the average values. The factory calibration Geo Exploration (P/N 730-0187) was used for the analysis of the stone tools due to the low or absent Sn concentrations on these materials. The factory calibration "Geomining (P-N 730.0203)" was used for the cassiterite samples analyses, since this calibration has a higher accuracy for high concentrations of ore metals such as Sn, Fe and Au (Bruker 2019).

### 2.4 WDXRF

Wavelength Dispersive X-Ray Fluorescence (WDXRF) chemical analyses were performed on the cassiterite samples from the Geosciences Museum of the IST (Lisbon, Portugal) and fieldwork, using a PANalytical XRF-WDS 4 kW AXIOS (PANalytical B.V., Almelo, The Netherlands) sequential spectrometer (Rh X-ray tube). The samples were analysed under vacuum without any preparation and a standardless semi-quantitative analysis was performed using the SuperQ software package (V.5.3A PANalytical B.V.). Details on the analytical methodology were already previously presented in the work Figueiredo et al. (2018).

### 2.5 SEM-EDS

Scanning electron microscopy (SEM) was performed on: one cassiterite sample collected during fieldwork; three cassiterite samples from the Geosciences Museum of the IST; and on some material collected from the surface of the stone tools from the Museu de Jazigos Minerais Portugueses (Porto, Portugal). The equipment used was a Zeiss DSM 962 (Carl Zeiss AG, Oberkochen, Germany) with a secondary electron detector (SE), a back-scattered electron detector (BSE) and an energy dispersion spectrometer (EDS) from Oxford instruments INCAx-sight (Oxford Instruments, HighWycombe, UK). The EDS system is equipped with an SDD detector, with PentaFET precision and a resolution of 125 eV at Mn K at 5,9 KeV, with the capacity to detect elements with atomic number superior to 5. The observations were performed using a working distance of 25 mm and an acceleration voltage of 20 kV. Quantitative analyses were performed based on the ZAF correction factors.

The collection of small size surface deposits on the stone tools was made using a (SEM-EDS conductive) adhesive carbon-tape, by peeling the stones' surfaces at specific locations, such as pounding facets or intents formed by grinding. All samples for SEM-EDS observation and analysis were prepared with a coating of Au.

### 2.6 Photogrammetric documentation

Images were acquired using a Nikon D3100, 24.2-megapixel, DSLR camera with AF-P DX NIKKOR 18-55mm f/3.5-5.6G VR Lens. Illumination was provided by 2x Hedler floodlights. White balance and colour reference Seculine ProDisk II were used for calibration purposes. RAW images were firstly processed to adjust white and colour balance, exposure, lens distortion and vignetting. The 3D processing was performed using Agisoft Metashape software V1.7.4 on a computer equipped with an AMD Ryzen 7 2700X Eight-Core Processor 3.70 GHz, 64 Gb RAM, Linux Mint OS.

## 3 Characterisation of the mineral resources

### 3.1 Geological context

The Ervedosa mine belongs to the Galicia Trás-os-Montes Zone (GTMZ), in the upper Subdomain of the Carracedo Structural Domain (CSD), on the eastern side of the Verin-Chaves-Régua-Penacova fault (VCRPF), where syn-tectonic granites, from the third deformation phase (D3) of the Variscan Orogeny, were emplaced in metasedimentary, volcanic-sedimentary and chalcosilicate parautochthonous sequences from the Upper Silurian age (Fig. 6).

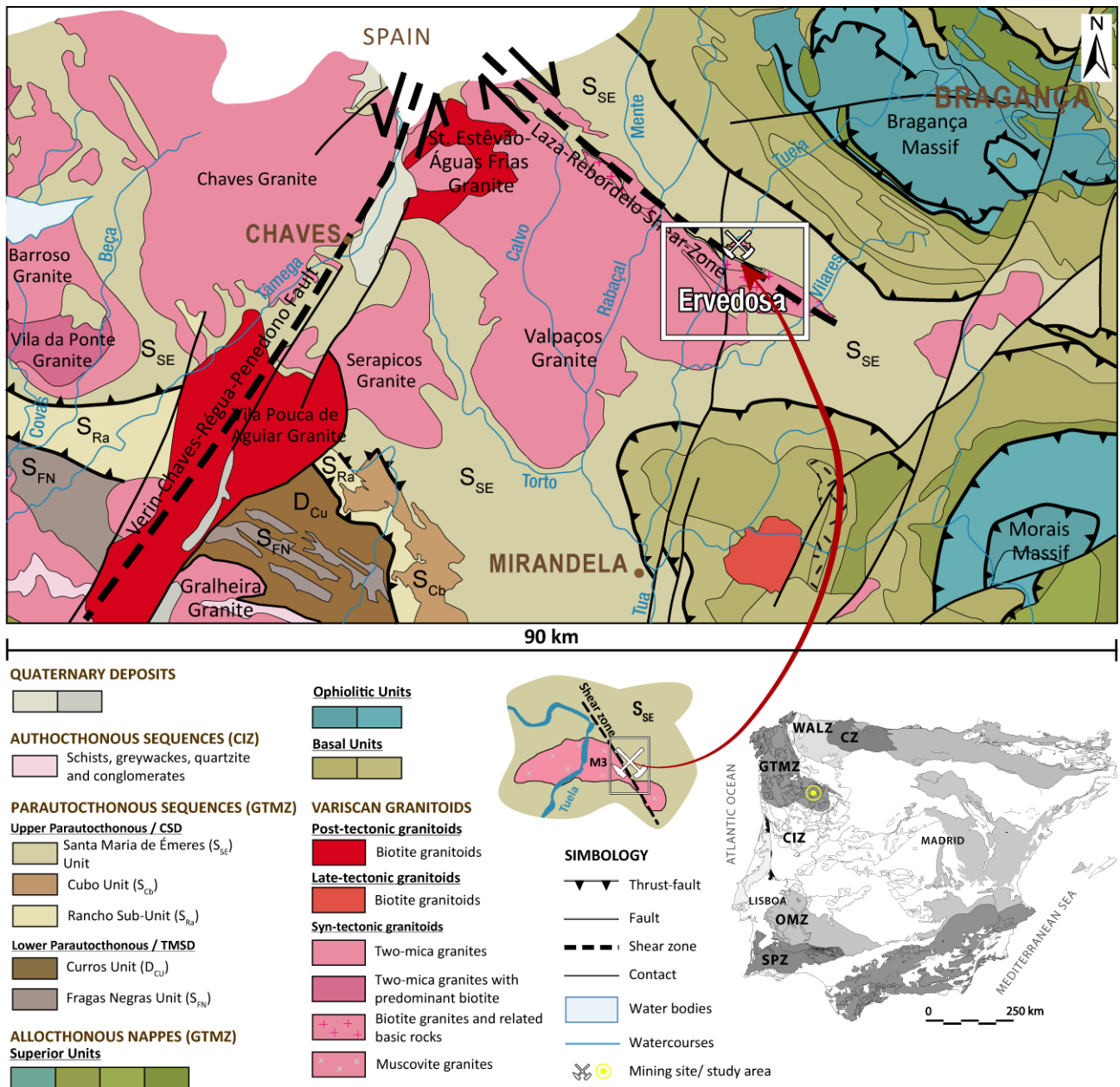
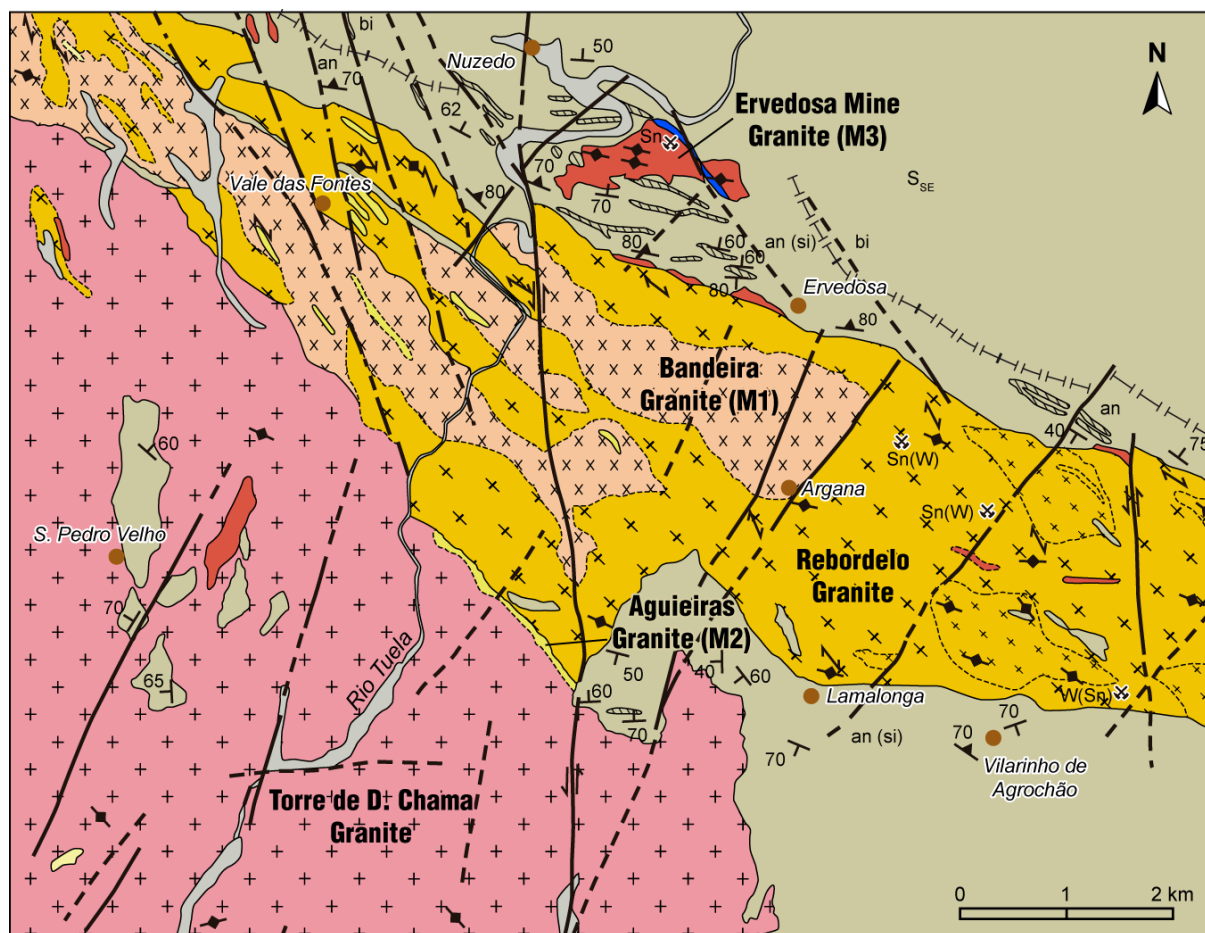


Fig. 6 – Geological context of the Ervedosa tin mine: (above) regional map with indication (white square) of the area represented in Fig. 7; (middle with red arrow) detail of the Muscovite granite zone of the Ervedosa mine, with indication of the area presented on Fig. 8; (bottom right) location of Ervedosa mine (yellow dot) in the western Iberian Peninsula geological zones. Images with adaptations and modifications following Rodríguez Fernández et al. 2015 and Gomes 1996.

The Variscan mountain ranges observed in the GTMZ are structures resultant from the Variscan Orogeny (between the Devonian and the Carboniferous ages) in a succession of three deformation phases (D1, D2 and D3). During the first two deformation phases (D1 and D2), the crustal thickening led to the genesis of synthetic granites (sin-D3) by anatexis of the crust. Later, a period of ductile-brittle and brittle deformation follows (late-D3 and post-D3), responsible for the fracture systems with NNE-SSW and NNW-SSE orientations that have concentrated important geological resources (Sant’Ovaia et al. 2011).

Ervedosa is depicted in the topographic military maps of Portugal (sheet 49 - Torre D. Chama, 1:25 000 scale) (Serviço Cartográfico do Exército 2013) and presented in the geological map of Portugal (sheet 2, 1: 200 000 scale) (Pereira 2000). Although this area is not covered by the 1:50 000 sheets of the geological map of

Portugal, the doctoral thesis of M. E. Gomes (1996) contains a detailed map of the geology of this region (Fig. 7) based on photogrammetry at 1:15 000 scale and military maps at a 1:25 000 scale.



**QUATERNARY DEPOSITS**

Conglomerates, sandstones, gravel, sand, silt and clay

**PARAUTOCHTHONOUS SEQUENCES (GTMZ)**

**Upper parautochthonous / CSD**

Santa Maria de Émeres (S<sub>SE</sub>) unit:

Feldspathic strips, black schists, and some litytes, phyllites, calcosilicate rocks, quartzite and some acid metavolcanic rocks

Folded quartzites

**GRANITOIDS**

**Late-Pos D3 granitoids**

Torre de D. Chama Granite:

Two-mica granite with medium to coarse grain size and porphyritic

**GRANITOIDS**

**Syn-D3 granitoids**

Ervedosa Mine Granite (M3):

Muscovite granite with fine to medium grain size

Agueiras Granite (M2):

Two-mica granite with medium to fine grain size

Bandeira Granite (M1):

Two-mica granite of medium grain size with a porphyritic tendency

**Pre-D3 granitoids**

Rebordelo Granitoide:

Biotite granodiorite of fine to medium grain size and porphyritic/ biotitic-muscovitic granite, medium grain size and porphyritic

**VEINS**

Ervedosa quartz vein

**Symbology:**

Stratification

Schistosity

Foliation

Shear zone

Contact

Likely contact

Fault

Likely fault

Likely metamorphic isograde

si (silmanite)

an (andalucite)

bi (biotite)

Old mining operations

Fig. 7. Detailed geology of the Ervedosa tin mine area showing the Ervedosa quartz vein (in blue) crossing the muscovite granite of fine to medium grain size (Ervedosa mine granite M3, in red) which is emplaced in metasedimentary, volcanic-sedimentary and calcosilicate parautochthonous sequences from the Upper Silurian age (Santa Maria de Émeres S<sub>SE</sub> unit, in grey-brown). Adapted from Gomes 1996 and updated with data from Gomes and Neiva 2002.

The tin exploited in the Ervedosa tin mine occurs as cassiterite minerals from hydrothermal quartz veins. According to Gomes and Neiva (2002), these veins resulted from the fractionation of a melt containing some

Sn, which initially crystallized in the form of two micas granites (syn-D3), then muscovite granites (syn-D3) and lastly quartz veins (post-D3) with significant concentrations of Sn. The erosion of these veins resulted in a concentration of cassiterite in the Tuela alluvium deposits. Therefore, Sn was mined both from the quartz veins and from the alluvium deposits (Gomes 1996). According to Inverno et al (2020), all other Sn occurrences registered in the Tuela river course are downstream from the Ervedosa mine. However, we cannot exclude that some of the cassiterite found in the local alluvium deposits were brought by the Tuela river from an unidentified cassiterite source upstream Ervedosa, and mined together with the sediments produced by the erosion of the Ervedosa veins.

According to Gomes and Neiva (2002), the first granite to crystallize in the area was the Bandeira granite, called M1. This granite is a two-mica granite, of medium grain size, slightly porphyritic, highly deformed and with low quantities of Sn, mainly found on biotite (Gomes and Neiva 2002).

The second granite to crystallize was most likely the Aguieiras granite, called M2. This is also a two micas granite that sporadically outcrops along this area and can be genetically related to the M1 granite. However, the M2 granite is more evolved, has a finer grain and is less deformed than the M1 granite.

The third granite to crystallize, and the last before the crystallization of the quartz veins, is assumed to be the granite of the Ervedosa Mine, called M3. This is a muscovite granite, with fine to medium grain size, with little deformation, and high content of Sn, compared to the other granites. Most of the Sn is found on the muscovites, but some also occurs as microscopic cassiterites, detected with a Scanning Electron Microscope (Gomes and Neiva 2002). Although the M3 granite is little deformed, it has an intense deformation on the eastern side (in the area of the mine) where it is displaced by a right shear zone (approximately N160°) cross-cutting both the granite and the sediments of the Santa Maria de Émeres (S<sub>SE</sub>), where it is emplaced (Gomes 1996). This shear zone played an important role in controlling the circulation of the hydrothermal fluids rich in Sn that later formed the quartz veins. There is one quartz vein emplaced along this shear zone called "Balbiani" during the 20th century mining operations. It is one of the largest veins containing Sn inside the mine, with around one meter width (Fig. 8), but not the richest. However, this vein is important due to its several branches of small quartz veins (of several centimeters in width) with high concentration of cassiterite. These branches formed along shear cracks (Gomes and Favas 2006).

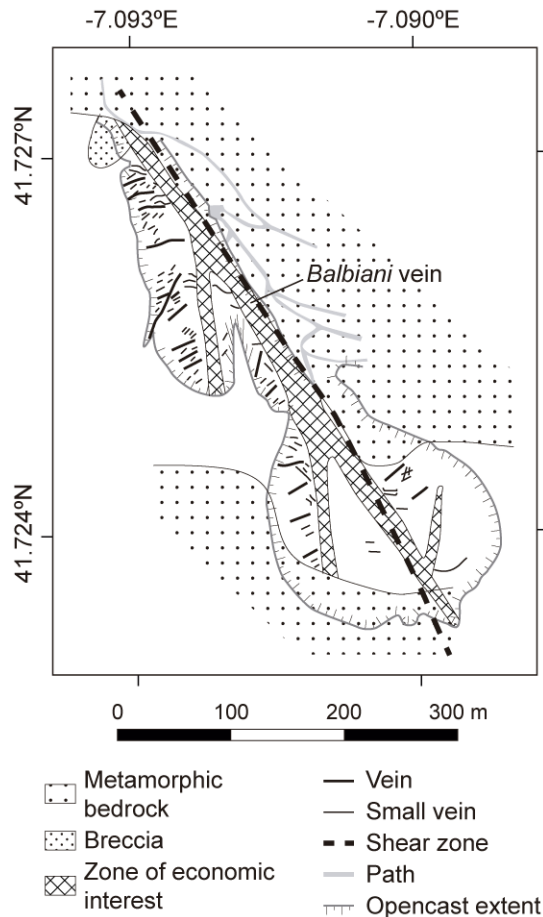


Fig. 8. Sketch of the mined area of the Ervedosa tin mine with depiction of the location of one of the largest quartz veins of metric dimensions (Balbiani vein) as well as several branches of small quartz veins, with directions of NS and NNE-SSW, with high concentrations of cassiterite (modified from Gomes 1996).

When Sn, as well as arsenic (As), was mined in Ervedosa, this type of small veins was the most sought for. Besides quartz ( $\text{SiO}_2$ ) and cassiterite, they could contain muscovite ( $\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH},\text{F})_2$ ), arsenopyrite ( $\text{FeAsS}$ ), sphalerite ( $(\text{Zn},\text{Fe})\text{S}$ ), pyrrhotite ( $\text{Fe}^{2+}_{0.95}\text{S}$ ), stannite ( $\text{Cu}_2\text{FeSnS}_4$ ), apatite ( $\text{Ca}_5(\text{PO}_4)_3(\text{OH},\text{F},\text{Cl})$ ), fluorite ( $\text{CaF}_2$ ), columbotantalite ( $\text{Fe}^{2+}(\text{Nb},\text{Ta})_2\text{O}_6$ ) and bismuth (Bi) (Gomes and Favas, 2006). Euhedral cassiterite crystals are very rare. Cassiterite is usually dispersed inside the veins, with dimensions from a few mm to round masses of up to 10 cm in diameter. Also, these Sn-rich quartz veins were usually crosscut by younger barren veins with milky quartz (Gomes 1996; Gomes and Farinha Ramos 2018; Gomes et al. 2008). Mining geological reports also refer the existence of very rich areas of the mine with small veins but with very regular ore content, and mostly related to greisen units (Anonymous 1914).

### 3.2 Analyses of cassiterite samples

Results of the pXRF and WDXRF analyses of the cassiterite samples are presented in Table 1 and Table 2, respectively (the pXRF spectra corresponding to each cassiterite sample can be found as online resource 1). The results show the detection of many elements besides  $\text{SnO}_2$  which are a result of associated minerals, mineral exsolution, inclusions or elements present as substitution of tin in the cassiterite lattice. Previous geochemical studies to cassiterite samples from the Ervedosa mine (Gomes and Favas 2006) have shown that cassiterite crystals exhibit oscillatory narrow and parallel zoning with darker zones exhibiting higher Ta, Nb, Fe and Ti contents (typically they sum up to  $<1$  wt.%) than the lighter zones, which are nearly pure  $\text{SnO}_2$ . Exsolution minerals are commonly columbite ( $(\text{Fe},\text{Mn})(\text{Nb},\text{Ta})\text{O}_6$ , ranging from Fe-columbite to Mn-columbite) and ixiolite ( $(\text{Ta},\text{Nb},\text{Sn},\text{Fe},\text{Mn})_4\text{O}_8$  or  $(\text{Ta},\text{Mn},\text{Nb})\text{O}_2$ , ranging from titanian ixiolite,  $\text{W}_{\geq}\text{Ti}$ -ixiolite and rarely niobian rutile), with ilmenite ( $\text{FeTiO}_2$ ) and wolframite ( $(\text{Fe},\text{Mn})\text{WO}_4$ ) mainly found in the darker zones of the

cassiterite. As mentioned before, besides cassiterite other primary minerals in the Ervedosa mine may be arsenopyrite and to some extent stannite. Arsenopyrite is the most abundant sulphide and contains inclusions of pyrrhotite, native Bi, bismuthinite ( $\text{Bi}_2\text{S}_3$ ) and matildite ( $\text{AgBiS}_2$ , with some additional Fe and Pb replacing Ag). Other sulphides are pyrite ( $\text{FeS}_2$ ), sphalerite ( $\text{ZnS}$ ) and chalcopyrite ( $\text{CuFeS}_2$ ).

Table 1. Results of pXRF analyses of cassiterite ore samples from Ervedosa at the Museu de Jazigos Minerais Portugueses.

Sample	source	$\text{Al}_2\text{O}_3$	$\text{SiO}_2$	$\text{P}_2\text{O}_5$	$\text{SO}_3$	$\text{TiO}_2$	$\text{Fe}_2\text{O}_3$	$\text{CuO}$	$\text{As}_2\text{O}_3$	$\text{SnO}_2$	Others
ERV-H	alluvial	5.7	15.2	n.d.	18.4	1.0	1.8	0.06	0.1	54.5	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{ZnO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$ ; $\text{WO}_3$
ERV-I	vein	1.9	13.0	n.d.	4.5	n.d.	0.7	n.d.	0.03	79.9	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$ ; $\text{WO}_3$ ; $\text{V}_2\text{O}_5$
ERV-J	vein	2.4	6.5	n.d.	4.9	0.3	0.6	0.3	n.d.	77.7	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{ZnO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$
ERV-K	alluvial	3.4	10.6	0.7	5.4	0.3	1.3	0.04	0.1	78.1	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$ ; $\text{WO}_3$
ERV-L	vein	1.5	7.5	0.8	2.1	0.3	0.3	0.2	n.d.	87.2	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$
ERV-M	alluvial	0.9	6.3	n.d.	3.0	0.2	15.6	n.d.	0.1	73.3	$\text{K}_2\text{O}$ ; $\text{CaO}$ ; $\text{Cl}$ ; $\text{ZnO}$ ; $\text{ZrO}_2$ ; $\text{Nb}_2\text{O}_5$ ; $\text{Ta}_2\text{O}_5$

Results in wt.% (normalised) presented in the form of pre-established oxides.

Table 2. Results of WDXRF analyses of cassiterite ore samples from Ervedosa from the Geosciences Museum at IST and the sample collected during field survey.

Sample	source	$\text{Al}_2\text{O}_3$	$\text{SiO}_2$	$\text{P}_2\text{O}_5$	$\text{SO}_3$	$\text{K}_2\text{O}$	$\text{CaO}$	$\text{TiO}_2$	$\text{Fe}_2\text{O}_3$	$\text{CuO}$	$\text{As}_2\text{O}_3$	$\text{Rb}_2\text{O}$	$\text{Nb}_2\text{O}_5$	$\text{SnO}_2$	$\text{Ta}_2\text{O}_5$	Others
B-029	vein	3.87	6.57	n.d.	0.59	0.38	0.58	n.d.	18.1	n.d.	0.62	n.d.	0.47	68.8	n.d.	
B-030	vein	23.7	30.5	n.d.	3.44	6.12	n.d.	0.20	19.5	n.d.	14.0	0.06	n.d.	2.45	n.d.	0.13 $\text{Bi}_2\text{O}_3$
B-031	vein	8.82	45.1	n.d.	14.4	1.99	n.d.	0.17	5.80	0.19	1.95	0.03	0.09	11.6	n.d.	9.87 $\text{ZnO}$
Field	vein	1.37	52.5	4.36	0.73	0.20	0.25	0.11	3.14	n.d.	0.11	n.d.	0.15	37.0	0.05	0.10 Cl

Results in wt.% (normalised) presented in the form of pre-established oxides.

All the samples analyzed in the present work (Table 1 and Table 2) show one or more of the combination of elements Ti, Nb and Ta (besides Fe), which are present as exsolutions such as columbite, ixiolite and ilmenite, or as substitution of tin in the cassiterite lattice. Other elements, such as Cu and/or As were detected in all samples and are most likely related to the presence of associated arsenopyrite and stannite minerals. The presence of Bi in the sample B-030 can relate to the presence of native Bi or bismuthinite and the relatively high amount of Zn and S in the sample B-031 can relate to the presence of sphalerite. The presence of elements such as Al, Si and K in all the analysed samples are most likely a result from associated gangue minerals in the samples (such as feldspar, muscovite and quartz).

SEM-EDS analyses of the samples B-029, B-030 and hand-sample collected in the fieldwork are shown in Fig. 9. The results show mainly relatively pure cassiterite crystals (taking into account the relatively high SEM-EDS detection limits), minerals with As and Fe that we interpret as weathered arsenopyrite since oxygen and carbon are frequently detected instead of sulphur (Murciego *et al.* 2011) and columbite or ixiolite micro-inclusions. Among the differences observed between samples, the microstructure of the field sample is more homogeneous comparing to the samples B-029 (with clear columbite and ixiolite micro-inclusions in the cassiterite) and B-030 (with high amounts of altered arsenopyrite).

Based on the present results, one can propose that the ancient use of the tin ores from Ervedosa to produce metallic tin resulted in a metal with trace elements such as Cu, As, Zn and Bi (As and Zn elements could be more affected by loss through volatilization than Cu and Bi). If the cassiterite was directly added to copper metal or copper ores to produce bronze (in a cementation or co-smelting process), one can assume that the presence of elements such as As in the bronze, which is frequently found in western Iberian bronzes at low content during the Bronze Age (frequently <0.5 wt.% As; Figueiredo *et al.* 2012; Figueiredo *et al.* 2016; Loureiro *et al.* 2016), would not necessarily mean that it (totally) originated from the copper ores, as normally assumed.

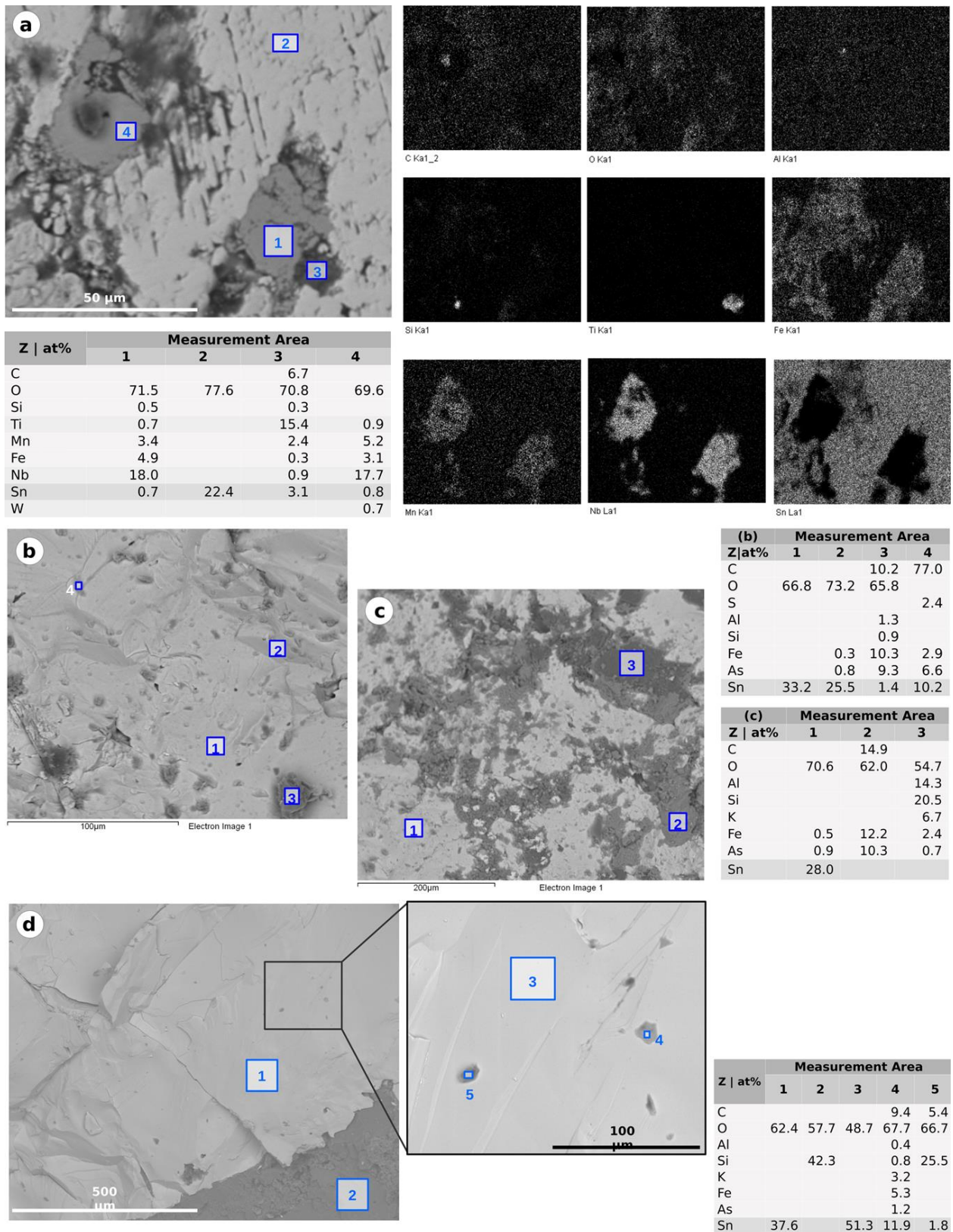


Fig. 9 – SEM-EDS observations of cassiterite samples: (a) sample B-029 from Geosciences Museum-IST, (b) and (c) B-030 sample from Geosciences Museum-IST, and (d) hand-sample collected during fieldwork at the Ervedosa mine. The interpretation of analyzed areas are as follow: a1 – columbite group mineral; a2 – cassiterite; a3 – niobian rutile; a4 - columbite group mineral with wolframite; b1 – cassiterite; b2-4 – altered (weathered) arsenopyrite within cassiterite; c1 – cassiterite; c2 - altered (weathered) arsenopyrite; c3 – K-feldspar or muscovite; d1 – cassiterite; d2 – quartz; d3 – zoned cassiterite; d4 – K-feldspar or muscovite with altered arsenopyrite; d5 – quartz.



## 4 Mining and ore processing stone tools

### 4.1 Description and interpretation use of the tools

According to Timberlake and Craddock (2013), Later Prehistoric mining and ore processing tools can be divided into three categories: hammers, pounding tools and crushing anvils. In the Ervedosa collection, we could identify one hammer (ERV-G), one pounding tool (ERV-B), two crushing anvils (ERV-A and D) and four tools that were used both as pounding tools and crushing anvils (ERV-C, E, F and E-01).

The hammer (ERV-G) is made of cobble stone. It is a double-ended mining hammer with a slight partial transversal groove and a small notch in the superior edge. It is partially broken, with missing flakes on the lower half, at both ends, resulting from its use for smashing rocks. Some blows' impacts are visible on the best-preserved end. The ERV-B item (Fig. 4 and online resource 3) was used as a pounding tool. It is an unmodified cobble stone, the second largest tool from the Ervedosa mine. Some pounding facets on both ends and a small broken flake on the widest end are visible. This kind of large crushing tool could be used held with both hands, without hafting (Timberlake, Craddock 2013: fig. 7c). Other items were used as pounding tools, also hand-held. The ERV-F and the E-01 tools show several pounding facets on their edges (Fig. 4 and online resource 7). The ERV-C piece also presents some impacts on one end and a small broken flake which could result from this use (fig. 4 and online resource 4). The ERV-E tool presents only a few impacts, but the broken flakes could be considered as resulting from pounding (fig. 4 and online resource 6).

The ERV-D tool seems to present a pounding facet only at one corner (Fig. 4 and online resource 5). In fact, this tool was used mainly as a crushing anvil, as indicated by the indents on the four main faces. These indents are quite small, between 2.5 and 3.5 cm in diameter, and around 1 cm in depth. We can observe such indents on ERV-C, ERV-E, ERV-F and the E-01 tools. Their diameter ranges from 2.5 to 4.5 cm, and they are all around 1 to 1.5 cm deep. This indicates that these tools had a double function, as pounding tools and as small crushing anvils. The indents are present on opposite faces for the ERV-F and the E-01 tools and on three to four faces on ERV-C, ERV-D and ERV-E tools (Fig. 4 and online resources 4, 5, 6, 7).

The largest piece, ERV-A, is a flat crushing anvil made of a piece of granite with three indents a little larger than on the other tools, as their diameter can reach 5.5 cm and they are close to 2 cm in depth (Fig. 4 and online resource 2). This block seems to have been taken from an outcropping granite or from the barren rocks extracted from the galleries. It was only used as a crushing anvil.

The small size of the indents observed on these tools indicates that the purpose was to crush rather small elements. We can also notice that they do not show intensive work: the impacts on pounding tools are quite reduced and even the flat anvil ERV-A only has three indents while its size would have allowed it to have more.

### 4.2 Rock-type, provenance and use vestiges of the stone tools

The compositional results of the pXRF semi-quantitative analyses of the stone tools from the Museu de Jazigos Minerais Portugueses are presented in Table 3. The pXRF spectra corresponding to each artefact can be found as online resource 9.

Table 3– Results of pXRF analyses to the stone tools from Ervedosa at the Museu de Jazigos Minerais Portugueses.

Sample	Type	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	SO <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	MnO	Others
ERV-A	granite	71.5	16.5	3.9	5.3	1.7	0.15	0.21	0.03	CaO
ERV-B	amphibolite	46.5	13.8	0.4	4.0	12.0	1.0	0.3	0.21	CaO
ERV-C	amphibolite	46.3	13.8	0.6	6.0	17.1	1.1	0.9	0.23	CaO
ERV-D	amphibolite	40.9	15.7	0.7	4.9	15.6	2.8	0.5	0.24	CaO
ERV-E	amphibolite	49.8	17.6	0.9	3.2	13.2	1.0	0.5	0.17	CaO
ERV-F	quartzite	71.2	6.6	0.42	17.6	1.6	0.08	0.49	0.17	CaO; MgO
ERV-G	dolerite	68.8	13.5	2.82	3.2	6.1	0.57	0.9	0.04	CaO; MgO

Results in wt.% (normalised) presented in the form of pre-established oxides

The semi-quantitative chemical composition of the granite processing tool (grinding stone ERV-A) is consistent with the published composition for tin-bearing granites from Ervedosa analysed by Gomes and Neiva (2002) and Neiva (2002). This may suggest that this tool originates from the Ervedosa granite.

The stone processing tools made of amphibolite (pounding stone ERV-B to ERV-E) are unlikely to be from Ervedosa, since the Ervedosa mine is in a granitic zone and amphibolites are from metamorphic zones associated to ultramafic rocks. Therefore, these materials are more prone to come from the Bragança Massif, with ultramafics terrains. The presence of these round-shaped, rolled amphibolites in Ervedosa can be explained by fluvial transport of eroded blocks along the Tuela river from the Bragança Massif, about 15 km northeast from the mine.

The quartzite processing tool ERV-F presents a chemical composition rich in  $\text{SiO}_2$ . This is likely a quartzite that outcrops in all the surrounding area of the mine, belonging to the "Parautochthonous Complex" (see, for instance, the quartzites from the nearby Vila Pouca de Aguiar area analysed by Aires et al. 2013).

The SEM-EDS analysis made to the collection of surface deposits on the stone tools allowed the detection of Sn-rich particles in three of the tools: the stone anvils ERV-A and ERV-E and the stone pounding tool ERV-B (Fig. 10). The Sn-rich particles are visible in several regions of the carbon-tape, as shown in BSE images and verified through the EDS micro-analysis. Some reservations about a possible intrinsic origin of the Sn-particles on the granitic surface of ERV-A anvil, of possible local origin, could be risen since it is known that small cassiterite crystals can occur occasionally in the granites from Ervedosa (Gomes e Neiva, 2002). However, the finding of these Sn-particles also on the surfaces of the two other tools (mortar/anvil ERV-B and stone pestle ERV-E), both fluvial-transported amphibolite, leave little doubt on their use for processing cassiterite ores or perhaps tin slags. With the present data it is not possible to distinguish between remnants of cassiterite ore or some other tin-related material, such as tin slags. Although the location of these tools in a tin mining site would primarily suggest ore processing, the detection of a Fe-rich micro-sphere on the ERV-A mortar surface (Fig. 10, measure a3) could rather suggest a slag remnant. Ancient slags are known to be highly heterogeneous at a micrometre scale, with the formation of various phases (Malham 2010). A small particle with a spheric shape could have formed in a liquid matrix of a metallurgical slag. Either way, these findings support the hypothesis of mining and processing of tin ores in Ervedosa mine during the Later Prehistoric period.

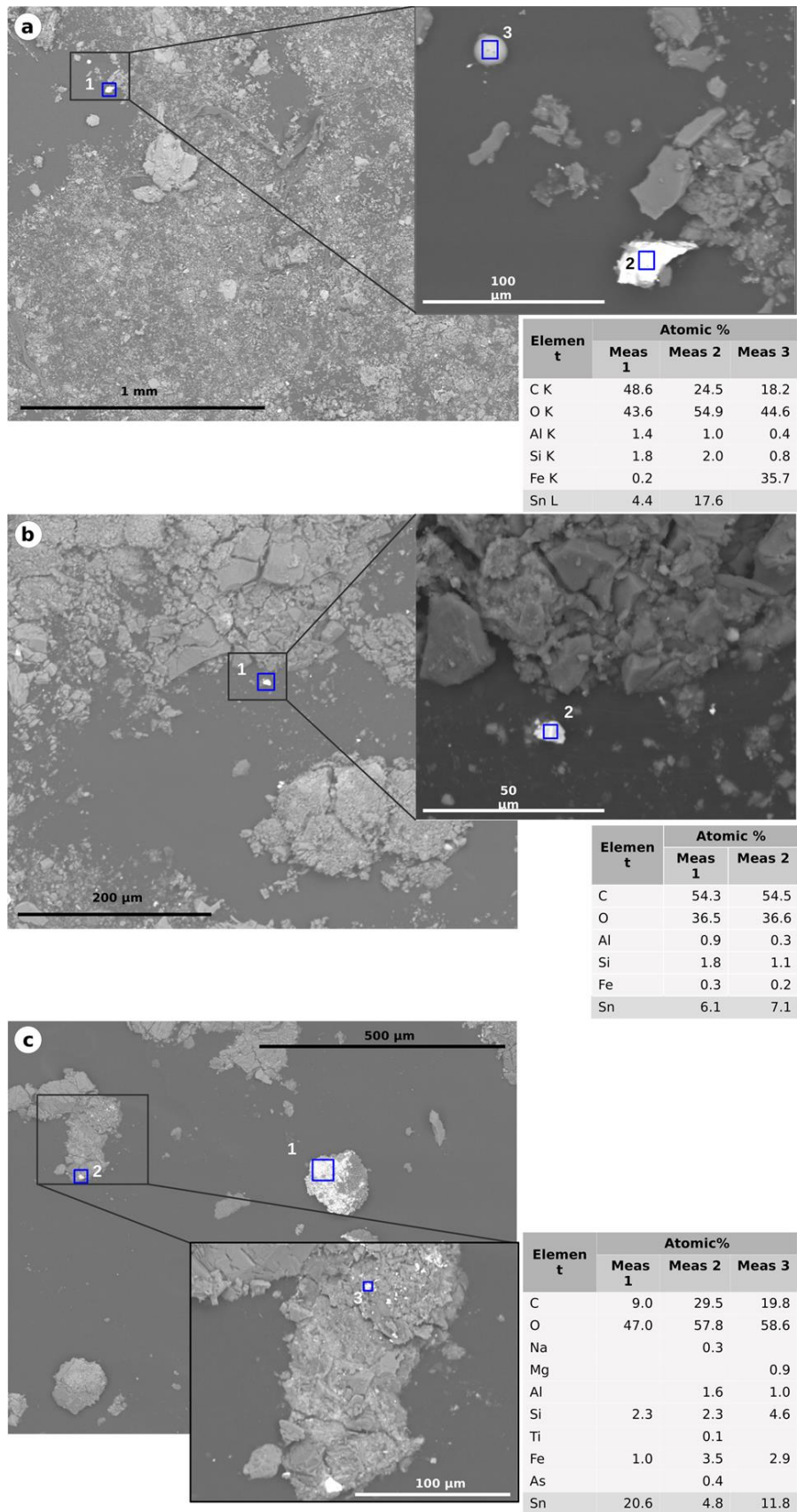


Fig. 10 – SEM-EDS observations (BSE images and results of EDS analysis) of the Sn-rich and one Fe-rich traces on the surfaces of three of the artefacts originating from Ervedosa (Museu dos Jazigos Minerais Portugueses): (a) ERV-A stone mortar/anvil, (b) ERV-B pounding tool, (c) ERV-E pounding tool/crushing anvil. All EDS analyses relate to Sn-rich particles, except for a3 that refers to a Fe-rich particle of spherical shape (note that very high C concentrations are due to the carbon tape where the particles are attached).

## 5 General discussion

The kind of cobble stone tools as those found in Ervedosa are known from Copper Age to Early Iron Age mines all over Europe (de Pascale 2003; Timberlake and Craddock 2013; O'Brien 2015). They were used before metallic tools became common, alone or associated with a previous fire-setting inside the mines, and for crushing ores or slags near the mines.

The stone hammer from Ervedosa is quite simple and is of a very common type used during the Chalcolithic (3<sup>rd</sup> millennium BCE) and Bronze Age (2<sup>nd</sup> millennium and early 1<sup>st</sup> millennium BCE), as evidenced by numerous findings in copper mines (Rafel et al. 2018: 76; de Pascale 2003). It corresponds to the double ended hammer described by Timberlake and Craddock (2013: 43), with notched edges and slight transversal groove, as some elements from Copa Hill mine. We can also compare it with the type 3a2 defined by M. Hunt Ortiz for Southwest Iberia (Hunt Ortiz 2003: 285). Some examples of notched hammer stones are reported in Austria, from the Bronze Age district of Mitterberg (O'Brien 2015: 172-173). It could also be compared with one from the Cerro Minado (Almería, Spain), dated in the late Chalcolithic period (Delgado-Raack et al. 2014: 30 and fig. V). This kind of hammer is also known in the Sierra del Aramo mines (Asturias, Spain), but with deeper grooves. The tools from the Asturian mines show significant reuse of broken tools (de Blas Cortina 2014: 66), which does not seem to be the case for the hammer from Ervedosa. The proximity between the mine of Ervedosa and the Tuela river, where the cobble stones came from, made them easily available so they could be discarded when broken.

Crushing tools like the ones found at Ervedosa are usually found near the mines or in settlements where metallurgical activities took place, sometimes somewhat distant from the mining site. They are related with the concentration of ore, to separate the sterile gangue from the ore itself, with the crushing of ore before metallurgical processing and with the crushing of slags to pick the metallic prills trapped inside the slags (Hunt Ortiz 2003: 292; Hamon et al. 2009: 199-201; Timberlake and Craddock 2013: 45). Most of the artefacts from the Ervedosa mine are cobble stones presenting marks of use on the edges and some small depression on the flat faces. Many parallels can be found in European mining or metallurgical sites dated between the Chalcolithic and the Bronze Age (Hamon et al. 2009). In the Iberian Peninsula, we can refer to a similar artefact in the Solana del Bepo copper mine (Rafel et al. 2017: 148-149). This kind of indented area in the flat surface of the tools is also described in the small crushing anvils from Copa Hill, United Kingdom (Timberlake and Craddock 2013: 48). In the Cabrières copper mining and metallurgical district (France), these small indents on the flat surfaces are related with an intensive crushing of small elements. Analysis of their surface show that it was in some cases used to finely crush ore before smelting and in other ones to crush slags to obtain the copper trapped inside (Hamon et al. 2009: 197-199). The percussion marks on the edges of the same tools are related with their use as pounders, showing a double function of these tools (Hamon et al. 2009: 199).

The flat granitic stone with three small indents from Ervedosa (ERV-A) can rather be compared with large mortars. Some examples are also known from mines in Southwest Iberia. In the Ruy Gomes copper mine (Beja, Portugal), a flat anvil of this kind, used in upper and lower faces with one depression in each face, is reported among nearly 30 grooved hammerstones (Flores and Araújo 1945). These tools correspond to the type A mobile mortars defined by Hunt Ortiz (2003: 292-293).

We should however note that although several mining and processing tools are known in Iberia and Western Europe, they are mainly related to copper mines. Regarding tin mines, we are only aware of two other sites, one in Portugal and another in Spain, with Later Prehistoric mining and processing tools. From Portugal, we know of a stone hammer found at the Cabeço da Argemela in Fundão (central Portugal) and currently in exhibition at the archaeological museum of this town (Museu Arqueológico José Monteiro) (Marques et al. 2011-2012: 95; Vilaça et al. 2011: 433). This tool, made of granite, is much bigger than the tools from Ervedosa and has a spherical shape of ca. 21 cm in diameter (Fig. 11 and online resource 10). It has a complete transversal groove, but it is broken and only one half is preserved. The precise finding location of this hammer

was not registered. However, the settlement at the top of the Cabeço da Argemela hill had two main occupation phases, Late Bronze Age/Early Iron Age (ca. 1300/1200-900/800 BCE) and Late Iron Age (ca. 2nd-1st century BCE) (Vilaça et al. 2011: 428). Also, quartz veins bearing cassiterite are present 300 m to the northwest slope of the settlement. These veins were mined during different periods of the 20th century. Mining works between 1956 and 1961 produced 150 tons of SnO<sub>2</sub> (Bastos 2017). Although old mining works have not been archaeologically documented, the presence of this hammer at the site clearly relates to Later Prehistoric tin mining. Its large size and shape dictate its use, that was different from those in Ervedosa: this hammer would have been hung with a rope to a tripod in front of the rock and dropped against it to break it (Timberlake and Craddock 2013: 48). In respect to stone tools related with tin mining from Spain, we can refer to some small pounders, of spherical shape, found around the tin mine of San Cristóbal (Logrosán, Spain), dated between the end of 9th and the beginning of 6th century BCE. These tools, which show some resemblances in size and shape to the ones of Ervedosa, are related to ore concentration and crushing of ore (Rodríguez Díaz et al. 2013: 102).



Fig. 11: The stone hammer from the Cabeço da Argemela (picture by Pedro Mendonça, artefact nº MAMJM167).

Regarding precise chronological attributions based on mining and processing tools, different interpretations can be proposed depending on the type of objects and on historical-archaeological traditions. The use of the crushing tools is generally attributed to a larger time frame than the use of stone hammers, as they are present in archaeological sites from the Chalcolithic and in some cases up to Roman contexts (Hunt Ortiz 2003: 292-293). Their use in the Chalcolithic period (3<sup>rd</sup> millennium BCE), Bronze Age (2<sup>nd</sup> millennium BCE and early 1<sup>st</sup> millennium BCE) and Orientalizing period (ca. 900-550 BCE, period starting with the establishment of Phoenician colonies in the western coast of the Iberian Peninsula) is quite well demonstrated in Southwest Iberia by their association with other elements of clearer chronology and also by some archaeological excavations. Crushing tools used in Roman context appear in sites with a diachronic activity which started in the Bronze Age, as in Cerro Muriano or Rio Tinto (Hunt Ortiz 2003: 124-125 and 137). Some of the crushing tools considered to be Roman in Cerro Muriano are also much bigger than the ones known from clear Later Prehistoric contexts, so this typological change could be a sign of chronological difference (Hunt Ortiz 2003: 293). Furthermore, the Roman stone crushing tools from Tresminas gold mine (Trás-os-Montes, Portugal) are clearly different from the tools from Ervedosa, as they are squared blocks of bigger size, with deeper and larger hollows (Whal-Clerici 2020: 190 and 206). Based on these evidences, and whenever there is lack of clear stratigraphic contexts, small cobble stone tools as the ones from Ervedosa should not be considered to be of

a late chronology, such as Roman. In the British Isles, these kinds of tools are known in Bronze Age mining contexts, or Early Iron Age contexts at the latest (Timberlake and Craddock 2013: 45).

In Iberian Peninsula, the first tin bronze artefacts appear in Late Chalcolithic/Bell Beaker contexts (last half of the 3rd millennium to early 2nd millennium BCE), among arsenical copper artefacts. These findings have been found in different locations of the Peninsula, with examples in Guidoiro Areoso, Pontevedra, NW Spain and Bauma del Serrat del Pont, Gerona, NE Spain (Fernandez-Miranda et al., 1995). In the Trás-os-Montes region, the earliest known bronzes are plain bronze axes of the “Bujões/Barcelos” type, attributed to the Middle Bronze Age (ca. 1800-1300 BCE) which were found at Bujões (Vila Real). Analysis of these artefacts show that they have 10-11 wt.% Sn, ca. 0.1% Pb and ca. 0.3% As (Figueiredo et al., 2012).

The earliest bronze metallurgical operations so far identified in Trás-os-Montes region is attributed to the second quarter of the 2<sup>nd</sup> millennium BCE, in Fraga dos Corvos site (Macedo de Cavaleiros, Portugal). Here, a metallurgical foundry area was found with bronze nodules (prills with 10-14 wt.% Sn, <0.5 wt.% Pb and <0.1 wt.% As) together with one crucible fragment (with traces of Cu and Sn) and two stone mould fragments to cast awls (Senna-Martinez et al. 2011). These materials were associated to a cultural environment comprising manual pottery with epi-bell-beaker geometric comb-stamped motifs of “Cogeces” or “Protocogotas” type (Early/Middle Bronze Age, first half of second millennium BCE). In this site, a second moment of occupation, attributed to the Early Iron Age or Orientalizing period was also detected, with the presence of bronze artefacts with typologies which suggests a Phoenician origin, or, at least, a strong influence from the southern areas of the Iberian Peninsula (Figueiredo et al. 2009). These artefacts show that Mediterranean influences reached the north of Portugal during the first half of the first millennium BCE. Unfortunately, not many more metals have been studied from the Trás-os-Montes region due to a lack of dedicated archaeological excavations and subsequent archeometallurgical approaches. Exceptions worth of notice include the Moinhos das Golas collection, where numerous bronze and some brass artefacts were studied and identified as belonging to Bronze Age and possibly later periods (assuming that Mediterranean influences could have reached northern Portugal introducing brass during Orientalizing period; Loureiro et al., 2016). Of later periods, it is worth of mentioning the Late Iron Age Carvalhelhos hillfort (Boticas, Vila Real) where tin smelting activities have been identified (Figueiredo et al. 2018). Earlier examples, dated to the end of Chalcolithic and beginning of Bronze Age include the Carrapatas type halberds made of arsenical copper (9 examples found in Abreiro, Carrapatas, Vale Benfeito and Vimioso), which are in the origin of the Iberian Atlantic Bell Beaker or Early Bronze Age halberds typology (Garrido-Pena, 2022). Considering all these elements and the fact that bronze production is reported at least since the second quarter of the 2<sup>nd</sup> millennium BCE in the Trás-os-Montes region, we propose a chronology for the Ervedosa ancient cassiterite mining between the Middle Bronze Age and the Early Iron Age. Regarding the function of the tools, we propose ore crushing and concentration as most likely, but slag processing close to the mine cannot be completely discarded, as the Fe-rich micro-sphere found on the ERV-A mortar surface suggests.

Since some of the Ervedosa tools originated from around level 525 of the mine, more than 180 m above the level of the Tuela river, and since written and oral references mention ancient small galleries in the mountain slope, we can consider that there are strong indications for the mining of tin veins. Excluding the Cabeço de Argemela example presented in this work, this is the second example of tin mining in a primary context reported with some detail in the Iberian Peninsula, with the first example being the mine of Logrosán (Cáceres, Spain) (Meredith 1998; Rodríguez Díaz et al. 2019; Rodríguez Díaz et al. 2001). The identification and exploitation of primary cassiterite veins in pre-Roman times by the local communities does not mean that the secondary cassiterite deposits from the Tuela river were discarded as a tin source at the time. We want, however, to highlight that for tin procurement, as for copper, Later Prehistoric ore catchment included primary resources.

Regarding the regional archaeological context, the search for tin resources has been considered as a significant factor to explain the high density of small to medium Later Prehistoric up to Early Roman sites close to cassiterite deposits in the Trás-os-Montes region (Lemos, 1993: 170). The settlement pattern around the Ervedosa tin mine is consistent with this, with more than ten recorded places within an 8 km range (Fig. 12). The nearby settlements are mostly fortified sites taking advantage of dominant topography, with wall enclosures and ditches. Their chronology is poorly known since no archaeological excavations have been done, with only surface survey providing partial chrono-contextual data (Lemos 1993; Maciel 1993). During these surveys, local-type ceramic sherds have been found, most of them attributed to the Iron Age. In one site Bronze Age ceramics have also been found (Muradal) and on two other sites (Castelo de Ervedosa and Múrio do Crasto) other materials such as *tegulae* indicate a longer occupation until the Roman period. The open settlements of Cabeço and Horta de São Jorge have only provided Roman materials, and the Canelho da Castanheira site could have also been occupied in medieval times. Besides the settlements, the quartzite crest of the Serro de Penhas Juntas (just 4 km NO to Ervedosa mine) was described in the beginning of the 20th century as a place widely worked for tin mining purposes (Lopo 1902; Alves 1934: 539-540; Lemos 1993: 103). Nowadays, only the Buraco do Serro gallery is accessible, on the southeast edge of the crest, showing at its entrance the use of fire-setting. This gallery is currently under study by our team and, unlike the Ervedosa mine, no industrial mining was performed in this site.

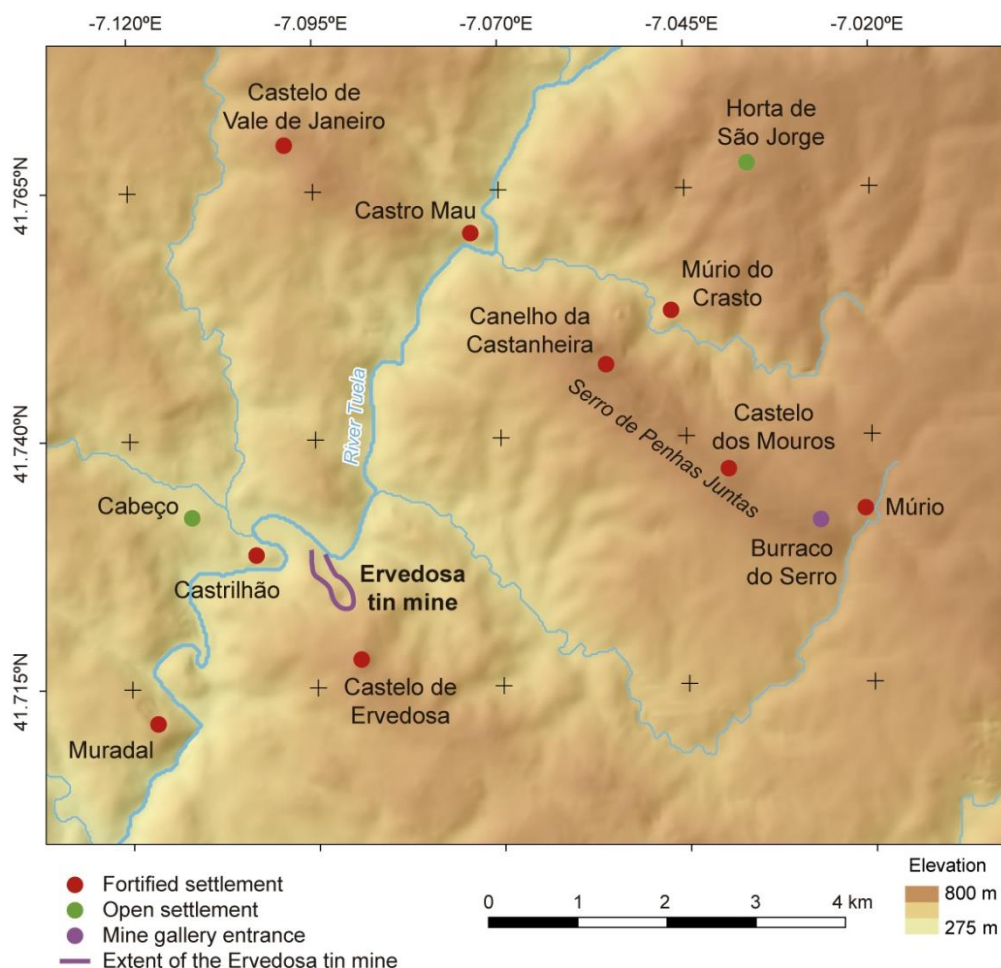


Fig. 12: The Ervedosa tin Mine and its archaeological context. Relief map based on EU-DEM v 1.1, Copernicus programme.

The possibility that local communities were taking advantage of numerous local tin outcrops, in a similar pattern that is assumed for copper mining in Iberia (Montero-Ruiz and Rovira-Llorens 2022), should be considered. In a 1937 doctoral thesis about the region of Trás-os-Montes, Virgílio Taborda brings together a

set of information, reflections and interpretative suggestions about the dynamics of development in the region. He records that “in the mineral deposits of Trás-os-Montes there are traces of very ancient exploitation, probably from the time of the Roman occupation. Throughout the Middle Ages there were probably also mining operations but only in the 15th and 16th centuries did documents begin to refer specifically to the Trás-os-Montes mines” (Fernandes, 2010). The chronology of these very ancient mines, actually unknown, could easily be in some cases Later Prehistoric, in the light of the data from Ervedosa.

The lack of archaeological excavations in the settlements surrounding the Ervedosa mine is an obstacle to a better understanding of the tin and bronze production activities in the area. The seemingly long-lasting local human presence between Bronze Age and Roman period may indicate that the cassiterite resources were sought at different moments, even if the stone tools studied here are better related to the earlier period. The mining activity discussed here, on the cassiterite-bearing veins, was likely a small-scale operation if compared to nowadays standards. The exploitation of alluvial tin in the Tuela river and its tributaries remains a complementary possibility for local populations to obtain this metal, as well as other tin outcrops existing in the region. The overall economic importance of local tin and bronze production during Middle Bronze Age to Early Iron Age (second quarter of 2nd millennium to first half of 1<sup>st</sup> millennium BCE) can only be assessed considering all these possibilities. In addition, future archaeological excavations in the settlements nearby the mine would be the best source of data to provide information about the ancient local economic activities. In the light of what has been reported for other Iberian places (Rodríguez Díaz et al. 2013; Figueiredo et al. 2018; Montes Landa et al. 2020; Figueiredo et al. 2022) we can expect to find more tools related to ore processing and metallurgical remains, such as tin slags or bronze debris.

## **6 Conclusions**

The mining and ore processing stone tools recovered at the Ervedosa mine are a good indication of Later Prehistoric tin mining, from a period between the Middle Bronze Age to the Early Iron Age, in a primary geological context. The presence of Sn-rich particles in the indents of the crushing tools is a significant element to confirm their use in tin-related processes.

Cassiterite samples from the Ervedosa site show the local abundance of tin resources in both primary and alluvial contexts. Regarding the chemical elements associated with the cassiterites ores and of metallurgical and archaeological interest, we highlight the presence of Ta, Nb and Ti (to a smaller extent W) that would end up in smelting slags, and the presence of As, Cu, Bi and Zn that could be incorporated as trace elements in the metal produced using the Ervedosa cassiterite ores.

In a wider perspective, the mining and processing tools from Ervedosa contribute to give further materiality to ancient tin mining in Western Iberian Peninsula, and provide evidence for tin mining in a primary context. This mine provides further evidence for tin and bronze production at Trás-os-Montes, where archaeological contexts for bronze production during the Middle Bronze Age and tin production during Late Iron Age had already been recorded.

Despite the archaeological importance of this site, the Ervedosa mine by itself is insufficient to explain the density of the settlement pattern observed in the surrounding area (up to 10 km) of the mine, neither can it be considered to be the single origin of tin supply at a regional (Trás-os-Montes) scale. Alluvium tin mining, whose importance is impossible to assess so far, and the existence of other mines in primary context that have not been studied yet, would also be part of the local economic framework. Future archaeological excavations are needed in the nearby settlements to collect data on local communities' engagement in tin and bronze manufacturing, to better understand their economies and cultural interactions during Late Prehistory.



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**Consent for publication:** All the authors agree for this paper to be published.

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**Code availability:** not applicable.

**Authors' contributions:** E.M., F.D., J.F., A.R., E.F., C.B. wrote the main manuscript text. Figures 1, 4, 5, 11 were prepared by E.M., figure 2 by J.F and E.M., figure 3 by E.M., A.R., figures 6-8 by F.D. and E.M., figures 9-10 by A.R. and E.F. Online resource 1 and 9 were prepared by A.R. and E.M., Online resources 2-8 by C.B. and E.M. Field work was performed by A.L., F.D., E.F., J.F., E.M. Sample selection was done by M.F.C.P., E.F., A.L. Chemical analyses and their interpretation were in charge of A.R., E.F., R.J.C.S., J.P.V., F.D., A.L. Photogrammetry of the tools was performed by C.B. Archive documentation was processed by E.M., J.F., F.D., A.L. All authors reviewed the manuscript.

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### **Captions for Online Resources:**

Online resource 1: pXRF spectra of six cassiterite samples from Ervedosa (Museu de Jazigos Minerais Portugueses) analysed. Legend: \* –  $K\alpha$  and  $K\beta$  lines of V, Cr and/or Mn overlap, uncertain presence of these elements; x – rhodium tube X-ray lines and other background peaks.

Online resource 2: 3D model of the tool ERV-A.

Online resource 3: 3D model of the tool ERV-B.

Online resource 4: 3D model of the tool ERV-C.

Online resource 5: 3D model of the tool ERV-D.

Online resource 6: 3D model of the tool ERV-E.

Online resource 7: 3D model of the tool ERV-F.

Online resource 8: 3D model of the tool E-01.

Online resource 9: pXRF spectra of six stone artefacts (ore processing tools) analysed from Ervedosa (Museu de Jazigos Minerais Portugueses).

Online resource 10: 3D model of the stone hammer from the cabeço da Argemela.