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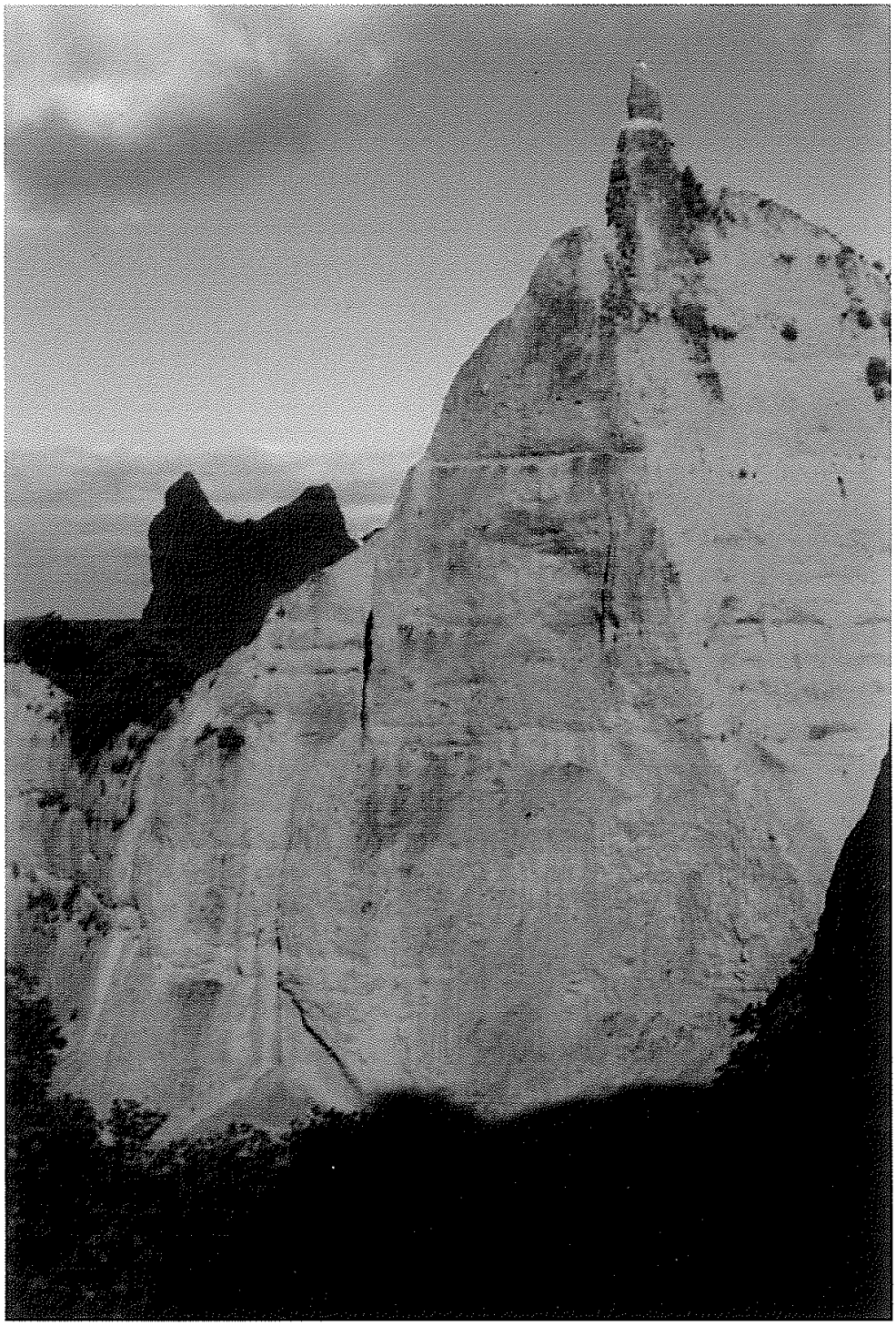

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ACTAS DEL QUINTO CONGRESO INTERNACIONAL SOBRE MINERÍA Y METALURGIA HISTÓRICAS EN EL SUROESTE EUROPEO (LEÓN – 2008)
LIBRO EN HOMENAJE A CLAUDE DOMERGUE

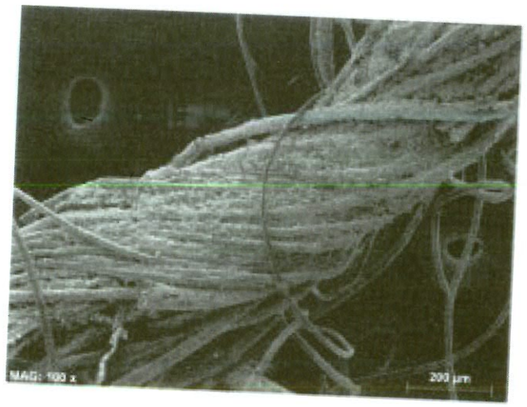

FIGURA 5. HILO DEL TEJIDO DE ARDITURRI. GROSOS DEL HILO SIMPLE DE TORSIÓN Z (TRAMA): 450`8 MICRÓMETROS. ÁNGULO DE TORSIÓN DE 58º. FOTO MEB. C. ALFARO

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ROMAN SLAG DISTRIBUTION IN THE PORTUGUESE SECTOR OF THE IBERIAN PYRITE BELT

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ABSTRACT

Detailed geological and mining mapping of Caveira, Aljustrel and São Domingos (Cu, Pb, and Zn-bearing pyrite masses) and of Fortes (Cu-rich lodes) allowed a better understanding of the Roman slag distribution at each site. Remnants of *in situ* Roman slag piles were identified, sometimes including pottery fragments. Locally, the Roman slag piles are covered by modern mining waste and have already been used for road paving and building. Using a CAD system, the results of the survey performed were compared with the oldest XIX century mine maps available for Aljustrel and São Domingos. The results indicate that Roman slag covers at present a total area of 200736 m^2: 78852 m^2 at Caveira, 79809 m^2 at Aljustrel, 28173 m^2 at São Domingos, and 13902 m^2 at Fortes; the XIX century maps show areas of 442424 m^2 at Aljustrel and 92410 m^2 at São Domingos. Taking the average slag density and thickness to be 3.1 $g.cm^{-3}$ and 3 m (2 m at Fortes mine), as suggested by field data, a total volume of 1.87 Mm^3 (5.79 Mt) of Roman slag is known to have existed in the IPB Portuguese sector. No remnants of Roman slag piles were so far found at Chança, Lousal and Montinho, also exploited in Roman times. Considering the efficiency values quoted in the literature as typical for Roman metallurgical processes and the slag mass estimated in the present work, a minimum of 370000 t of Cu, 139 t of Ag and 9 t of Au were produced in the Roman *Lusitania* Province, if conservative ore grades are assumed.

Key-words: Roman mining, Slag, Iberian Pyrite Belt, Caveira, Aljustrel, São Domingos, Fortes.

RESUMO

A cartografia geológica e mineira detalhada das áreas de Caveira, Aljustrel e São Domingos (sulfuretos maciços ricos em Cu, Pb e Zn) e da mina de Fortes (estrutura filoniana cuprífera) permitiu conhecer melhor a distribuição dos escoriais romanos em cada local. Nestes levantamentos assinalaram-se diversos escoriais romanos *in situ*, por vezes incluindo fragmentos de cerâmica. As escombreiras romanas encontram-se localmente cobertas por resíduos mineiros modernos, tendo sofrido em certos casos forte remoção, devido ao seu uso como material de construção e de pavimentação de caminhos. A cartografia realizada foi digitalizada em sistema CAD georreferenciado e comparada com a documentação cartográfica do séc. XIX disponível em escala de detalhe para as minas de Aljustrel e São Domingos. Os resultados indicam que as escórias romanas se distribuem actualmente por uma área total de 200736 m^2: 78852 m^2 em Caveira, 79809 m^2 em Aljustrel, 28173 m^2 em São Domingos e 13902 m^2 em Fortes; os mapas do séc. XIX revelam áreas de 442424 m^2 em Aljustrel e 92410 m^2 em São Domingos. Considerando uma densidade média de 3.1 $g.cm^{-3}$ para as escórias romanas e uma espessura de 3 m para os escoriais (2 m na mina de Fortes), os dados sugerem a existência de um volume total de escórias romanas de 1.87 Mm^3 (5.79 Mt) no sector português da Faixa Piritosa Ibérica. Durante os trabalhos de campo não se identificaram escoriais romanos nas minas de Chança, Lousal e Montinho, também exploradas em período Romano. Considerando a eficiência dos processos metalúrgicos romanos referenciados na literatura e assumindo o volume de escórias agora definido, bem como valores conservadores para os teores dos minérios explorados, estima-se um valor mínimo de produção de 370000 t de Cu, 139 t de Ag e 9 t de Au na província romana de *Lusitania*.

Palavras-chave: Mineração romana, escórias, Faixa Piritosa Ibérica, Caveira, Aljustrel, São Domingos, Fortes.

1. Introduction to the Roman mining in the Iberian Pyrite Belt Portuguese sector

During Roman times, several outcropping massive sulphide deposits located in the Portuguese sector of the Iberian Pyrite Belt (IPB) were intensely exploited. From NW to SE, these are Caveira, Lousal, Montinho, Aljustrel, São Domingos and Chança. Maximum known depth of Roman exploitation is 118 m at Algares, Aljustrel (the Roman *Vipasca*). These pyrite mines and other similar sites located in Spain (e.g. Río Tinto, Tharsis and La Zarza) were one of the most important base metal sources of the Roman Empire (Ortigão 1983, Domergue 2002). The metals left the Portuguese IPB sector by ship. Indirect archaeological remains point to a Roman road linking Aljustrel to the Guadiana River in the area of Mértola (*Myrtilis*), and Alcácer do Sal (*Salacia*) was also a probable export harbour, at least for the Caveira and Lousal production. Roman mining practice was aimed at the exploitation of secondary ores (oxidation zone – iron hat and supergene alteration zone) and, accessory, of primary pyrite ore; the ore was roasted in place to extract the metals for export. Some of the Roman mines were very big endeavours, which were directly under imperial or provincial control, and caused the first events of acid mine drainage known in Portugal (Matos and Martins 2006). Roman slag accumulations are present in several IPB mines, and can have considerable size (e.g. Aljustrel, São Domingos and Caveira in Portugal and Río Tinto, La Zarza and Tharsis in Spain). Some of it is covered by modern mine waste produced during the 19th-20th centuries (e.g. Mateus et al. in press). This study presents a global analysis of the Roman slag distribution in the IPB Portuguese sector, one of the mining wastes classes commonly considered in this mining province and a better indicator of the total ore processed at each site than the (often fragmentary) direct mining remains (e. g., shafts and galleries).

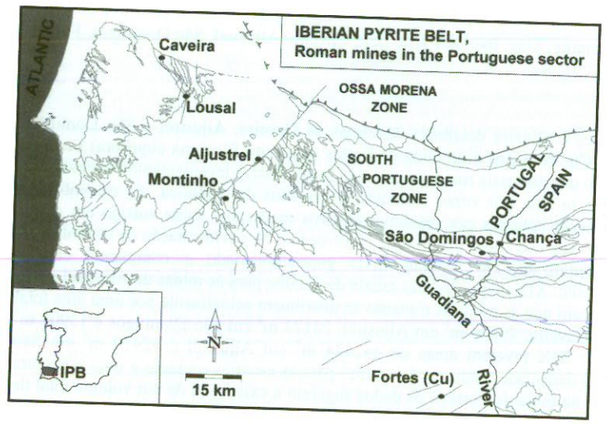

Fig. 1 – Roman mines in the Portuguese sector of the Iberian Pyrite Belt. Geological background adapted from Oliveira et al. 2006, Matos & Martins 2006.

2. Roman slag study methodology

Roman slag accumulations and other Roman remains were identified and mapped (1/2500 scale) as part of a detailed mapping program of the abandoned mining centres of the Portuguese IPB (Fig. 2); field mapping, aerial photograph and satellite images were used to improve survey quality. Mapping data were introduced in an AUTOCAD computer system, which was subsequently used to determine the area extent of each type of waste. The maps produced were compared to 19th century mine maps older than the beginning of modern mining activities, available for Aljustrel and São Domingos, respectively the 1867 Trastagana Company map (LNEG Archive), and the Carlos Ribeiro 1857 map (see Custódio 1996, 1999). In these two mines and at Caveira, the slag was characterised by multielemental chemical analysis and petrographic examination; where preservation is good, as at Caveira and São Domingos, image analysis, X-ray diffraction, INAA and ICP-MS chemical analyses after total acid digestion, and electron and particle-induced X-ray emission microanalyses (electron and micropixe analyses), were also used.

3. The Caveira Roman mine

The Caveira mine is located on an antiform structure with a nucleus of Devonian Phyllite-Quartzite Group (PQ) rocks surrounded by an Upper Famenian-Upper Visean sequence, the so-called Volcano-Sedimentary Complex (VSC), consisting of felsic metavolcanics, dark grey shales, and late metadiabases (Oliveira et al. 2006, Matos 2006). Several massive sulphide <10m thick lenses occur near the contact VSC/PQ, forming two main ore horizons, the first one comprising the NW-S. João and Salvador-Esperança lenses (Helena Shaft – western sector) and the second one the Canal-Frederico-Francisco and Augusto-António (Luísa Shaft – eastern sector) lenses. According to the 19th century description of the mining works (Cabral 1889), Roman mining remains consisted of several shafts and large volume of slag deposits (300000t estimated). Both isolated exploration shafts and twin shafts were also mentioned, especially from areas with underground waters.

Three mine water discharge galleries were found, one of them 1 km long. The main Roman mining areas were obliterated by the 19th century open pit. Despite of this, the Caveira mine is today one of the best IPB sites to observe Roman mining works. Slag deposits, more than 10 tunnels (<1 m^2section) and at least one mine chamber located at the Salvador open pit (western mine sector, see Figs. 2 and 3) can still be seen. The tunnels are present all over the Salvador pit wall (height 15 m) and were bored in rock wall mineralization host felsic well cleaved metavolcanics; they represent exploration tunnels probably used to search for stockwork/fissural mineralized structures, similar those described for São Domingos (see below).

The slag piles are located up to 670m northward and 570 m eastward of the Salvador orebody and consist of a small *in situ* accumulation at the right bank of the main tailing stream, covering a total area of 78852 m^2 (see Table 2). Since the 20th century this slag material is being extracted, mainly from the northern waste area, and reused for road paving. The slag consists of a glassy matrix with zoned, sometimes lath-like, fayalite and magnetite as the main mineral phases. Sulphides appear as recrystalized droplets, now consisting of anomalous pyrite/pyrrhotite, chalcopyrite and several non-stoichiometric ill-defined phases interpreted to be monosulphide liquid chilled before complete exsolution of the normal mineral phases could occur. The larger droplets show small grains of galena and of what seems to be some Pb alloy. Weathering is evident under the microscope as coatings (both external and inside voids and fractures) of hematite/goethite (maybe also lepidocrocite) and sometimes pyrolusite; the sulphide phases whether to covellite.

4. The Aljustrel (*Vipasca*) Roman mine

The Aljustrel mine is one of the biggest Roman mining sites in Portugal. The Roman mining regulations for this site are partially preserved in two bronze tables found in 1876 and 1906 at Aljustrel and archaeological excavations promoted around the Algares mine allow a good understanding of the *Vipasca* Roman mining site (see Viana et al. 1954, Alarcão 1983, Domergue 1983, 2002, Martins 1996, Gaspar 1998, Martins et al. 2003, Macias et al. 2008). Two outcrop massive sulphide deposits at Algares and S. João were mined for copper between the 1st and 3rd centuries A.C. (Alarcão 1983).

The mining exploitation at S. João is nowadays represented by small tunnels observed in the open pit western sector, developed along fault zones associated with sulphate veins (natrojarosite $NaFe_3(SO_4)_2(OH)_6$ and jarosite $KFe_3(SO_4)_2(OH)_6$, e.g. Matos et al. 2003, Fig. 3). No slag has ever been identified at S. João; probably the ore exploited in this secondary mine area was carried ~2 km up-stream to the Algares main mine sector. This area was dedicated to the exploitation of the Algares West, Central and East massive sulphide deposits. The former is the main mineralized structure and outcrops for ~900 m from the present Aljustrel town to the Água Forte stream (see Fig. 2); the deepest Roman mining works are 118 m below the surface (Viana et al. 1954). Roman remains at Aljustrel include: (*i*) several shafts and tunnels (Fig. 3) at the Algares West iron hat; (*ii*) house infrastructures at Transtagana/Procurador House; (*iii*) metallurgic infrastructures at Feitais valley; (*iv*) the Valdoca Roman necropolis; (*v*) large volume of slag; and (*vi*) numerous artefacts housed in several Portuguese and foreign museums (Martins 1996, Domergue 2002, Martins et al. 2003, Macias et al. 2008). The *Vipasca* village was located in the western area of the Algares iron hat. Roman slag piles, extending for more than 1200 m (see Fig. 2), cover the left margin of the Água Forte stream, possibly due to Roman time favourable topography and wind conditions. The Aljustrel 1867 mining map (unidentified author, Transtagana Company) shows several mine shafts along the Algares West iron hat and a NW-SE mine water discharge tunnel; at the main slag deposits, slag Cu rich zones were also identified.

The only sector occupied in the right margin is the Feitais metallurgic facilities where cementation-type tanks were recently identified (see *Scavrarii* Roman miners specific activity dedicated to slag reworking, Macias et al. 2008). Near the present-day mine Palace (north Algares) there is an isolated slag deposit not well correlated with iron hat ore. Comparison of the 19th century map with the survey of Matos (2005) shows the extent of Roman slag reworking and reuse; Roman slag was used by the Aljustrel mine concessionary company to build the "Água Industrial" dam wall (BAI dam, Fig. 2), and most of the Roman waste is now covered by modern pyrite waste.

The results of the 2005 survey indicate a total Roman slag area of 79809 m^2, while the 1867 map shows an area of 442424 m^2 covered by Roman slag (Table 2). In situ and reworked Roman slag has significant Pb, Cu, Zn, Fe, (As), (Sb) grades (see Table 1). Algares slag preliminary petrography shows evidence of pyrite, rare chalcopyrite and sulphosalt grains interstitial to the silicate (fayalite)+magnetite matrix.

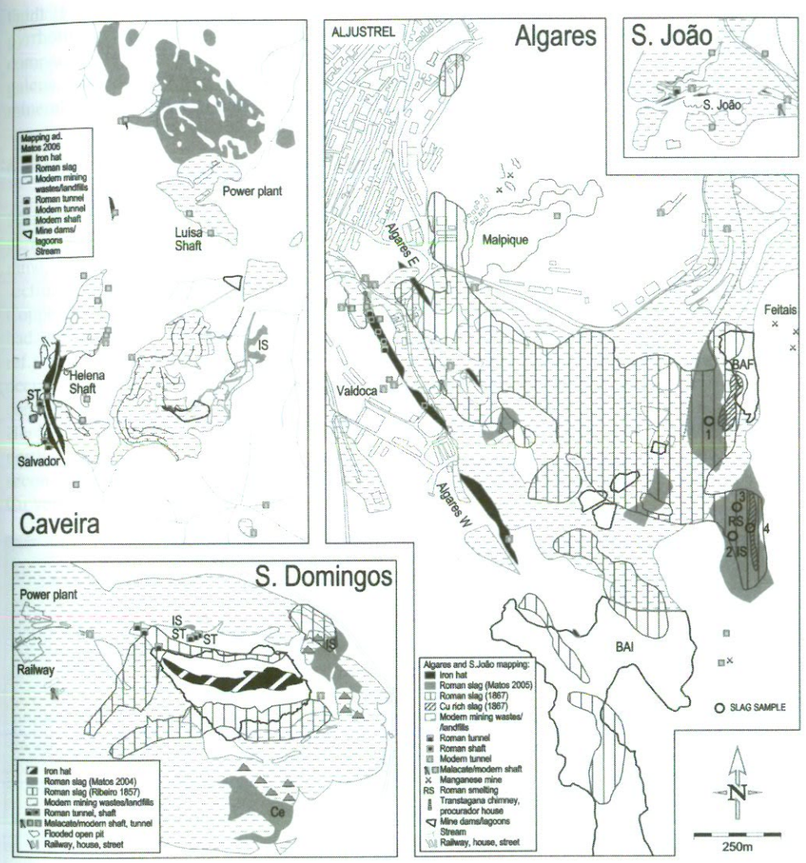

Fig. 2 – Roman slag at Caveira, São Domingos and Aljustrel (Algares and S. João) mines, adapted from Matos 2004, 2005, 2006 (DPMM INETI): **IS** – *In situ* Roman slag, **Ce** – pottery fragments; **ST** – Roman mining of stockwork-ore types. **Iron hats** indicate outcropping massive sulphide ore (estimated original extension at São Domingos). Triangles at São Domingos refer to Roman slag under 19th century mining waste (CONASA borehole data). Mining waste/landfills include dumps of pyrite ore, iron hat material, wall-rocks, modern slag and mine landfills. Aljustrel mine dams: BAI – clean water "Água Industrial" dam; BAF – acid water "Água Forte" dam. Other mine dams with acid waters.

Table 1 – Chemical ICP-MS ACME Lab analysis of Aljustrel (Algares) Roman slag. 1: thin slag, probably reworked in the 19th century; 2-4: in situ slag (location at in Fig. 2).

Slag samples		Algares 1	Algares 2	Algares 3	Algares 4
Ag	ppm	10	6	6	4
Al	%	0,88	1,43	1,42	1,25
As	ppm	1526	483	580	381
Au	ppb	77	26	23	18
Ba	ppm	77	157	190	201
Ca	%	0,02	0,05	0,02	0,03
Cu	ppm	2192	5536	4453	3311
Fe	%	13,82	15,10	15,51	7,77
Hg	ppm	15	1	4	3
K	%	0,30	0,29	0,23	0,15
Mg	%	0,16	0,15	0,20	0,11
Mn	ppm	202	354	280	749
Na	%	0,13	0,05	0,04	0,02
P	%	0,06	0,07	0,07	0,06
Pb	ppm	2621	3133	2533	2069
S	%	1,9	0,86	0,79	0,36
Sb	ppm	118	52	46	29
Se	ppm	20	3	6	3
Ti	%	0,01	0,02	0,01	0,01
Zn	ppm	358	645	410	269

5. The São Domingos Roman mine

The São Domingos pyrite orebody is located at the top of a VSC sequence, consisting of black shales, and felsic, basic and intermediate-basic metavolcanics (Matos et al. 2006, Oliveira et al. 2006). These rocks contact through thrust faults (vergent to SW) with shales and quartzites of the PQ Group and black shales of the Represa Formation, both of late Devonian age. Ore weathering originated an important W-E striking iron hat (500m long, up to 60m wide), intensely mined during Roman times and totally exploited between 1857 and 1966 (see Fig. 2). At São Domingos, the Roman mining works extend to >50 m depth (water pump wood wheels were mentioned in the 19th century, eight of them with 5,28 m diameter and two with 3,96 m diameter, Cabral et al. 1989). Mine mapping performed by Carlos Ribeiro in 1857 (see Custódio 1996, 1999) shows Roman slag scattered around the large iron hat that correspond to a differential erosion relief. By comparing the 19th century map with the modern survey (Matos 2004, Matos et al. 2006), slag transport and probable reworking during the 1850's and 1860's becomes evident. Nowadays, Roman mining remains consist of in situ slag accumulations located eastward (Fig. 3) and northward of the open pit, reworked slag southeast of the pit, several tunnels and great numbers of artefacts, e.g. a Roman wood wheel kept at the Musée National des Techniques, Paris (Custódio 1996, Domergue 2002).

Several pottery fragments were found in the slag piles. A CONASA (1991) mining waste evaluation program based on dump drilling, demonstrated that Roman slag is present under modern mining waste (see Matos 2004, Fig. 2). The 2004 survey indicates that, at São Domingos, the Roman slag occupies 28173 m^2 (see details in Álvarez-Valero et al. 2007, Mateus et al. in press); in the 19th century map, the Roman slag covers 92410 m^2 (see Table 2). Typically, the Roman slag is a glass-silicate aggregate with abundant oxides and some sulphides. The main silicates are zoned fayalite, melilite and pyroxene. Fayalite forms herringbone skeletal crystals near the original slag surface, but adopt a lath-like habit inside the slag, where it coexists with melilite and pyroxene. The main oxide is magnetite occurring as skeletal crystals in the glass-fayalite groundmass or as crystals associated to sulphide minerals;

the latter are sometimes shown to post-date magnetite. The main sulphides are pyrite and pyrrhotite, often intergrown with magnetite. Cu-rich phases normally similar to chalcopyrite are comparatively rare, but small aggregates of a Pb-rich assemblage, essentially, Pb metal and galena, sometimes with minor Pb-Sb-Sn sulphoantimonites are common; the main weathering minerals are iron (hydro)xides (Pinto et al., 2007, Mateus et al. in press).

6. Other IPB minor Roman mines: the Chança (Py) and Fortes (Cu) mines

Roman mining can be shown to have existed in other small IPB pyrite orebodies like Montinho and Chança. At Lousal mine, field data do not show any mining works from this period; nevertheless, partial exploitation of the South deposit cannot be excluded. The Chança mine, at the Portuguese/Spanish border has several Roman shafts (also twin shafts with ~1 m^2 section) in the iron hat hill. Related slag can be seen in the nearby Spanish Vuelta Falsa mine. Copper rich Late Variscan vein structures were also mined during Roman times. Some of them had mining activities since the Chalcolitic period (Alarcão 1983, Matos e Martins 2006, inc. ref.). One of the most important Roman slag accumulations related to this geological setting occurs at Fortes (Alcoutim, Algarve), where three Cu-rich, NE-SW veins were exploited, corresponding to 19th century abandoned mines Cerro das Pedras, Cerro do Forramerendas and Herdade da Malhada (Matos 2007). The Roman slag deposit covers an area of 13902 m^2 on the north bank of the Odeleite stream, ~500 m away from the ore vein. Pottery fragments and secondary copper minerals are visible. Although smaller than pyrite mines at IPB, the Roman Cu vein exploitation was also important due to the higher grades observed.

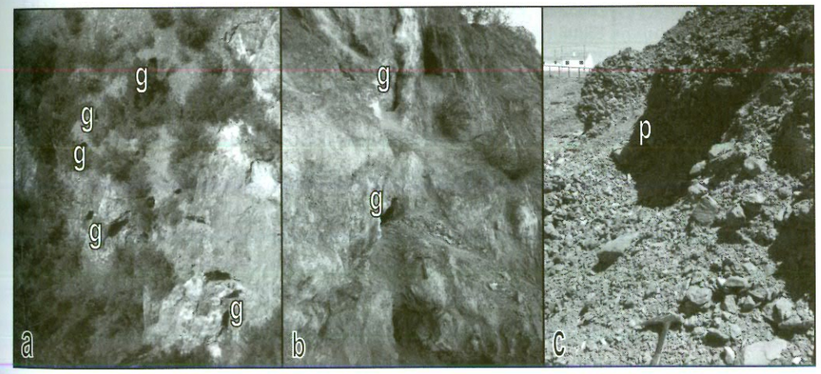

Fig. 3 – Roman mining works in the Portuguese IPB sector: a) – Roman galleries (g) at Salvador open pit (Caveira mine); b) – Roman galleries (g) at S. João open pit (Aljustrel mine) located in a fault zone with jarosite and hematite; c) – In situ Roman slag in the São Domingos eastern area, identified palaeo-soil (p).

7. Roman slag distribution in the southwest Iberian region

No remnants of Roman slag piles were found so far at Chança, Lousal and Montinho, all of them exploited in Roman times; at Chança, slag piles are found at the nearby spanish Vuelta Falsa mine. Table 2 shows a summary of the quantitative results pertaining to the Roman slag that can be extracted from the mapping data available. Taking the average slag density and thickness to be 3.1 $g.cm^{-3}$ and 3 m (2 m at Fortes mine) as suggested by field data, a minimum

total volume of 1.87 Mm^3 (5.79 Mt) of Roman slag is known to have existed in the IPB Portuguese sector (see Fig. 4). Even assuming these numbers reflect the total amount of Roman slag produced; the large scale of Roman mining operations is striking. It is generally assumed that the mass of slag equals the mass of processed ore. The exact grade of the oxidised ore exploited in Roman times is not precisely known, but is taken to be around 8% Cu in the IPB (Hong et al. 1996). The Roman metallurgical technology for Cu had at least 80% efficiency and so a minimum of 370000 t of Cu has been extracted from these south Portuguese mining centres, that is approximately 1000 t per year (see Raber 1987 and references therein for details on the calculation method and on the estimations it is based upon). There are, to our knowledge, no general estimates of the mean Ag grade in IPB Roman ores. The very Roman mining regulations for *Vipasca* suggest that the ore was extremely heterogeneous (entrepreneurs could bid for Cu or Ag veins). Geological considerations suggest 30 ppm Ag to be a fair estimate of the average grade of all the ore processed. This translates to 139 t Ag or an average production of 400 kg Ag per year for the whole Portuguese IPB. The same calculations, using an average Au grade of 2 ppm give a total production of 9 t Au (25 kg per year).

Table 2 – Roman slag distribution in the IPB Portuguese sector, tonnage and volumes calculated from total known areas determined from CAD mapping measurements.

Mine/Roman slag areas (m^2)	2004-2006 maps	19th century maps	Thickness (m)	Tonnage[1]	Volume (m^3)
São Domingos	28173	92410	3	859413	277230
Aljustrel	79809	442424	3	4114543	1327272
Caveira	78852	0	3	733324	236556
Fortes	13902	0	2	86192	27804
Total	**200736**			**5793472**	**1868862**

[1] d=3,1 gcm^{-3}

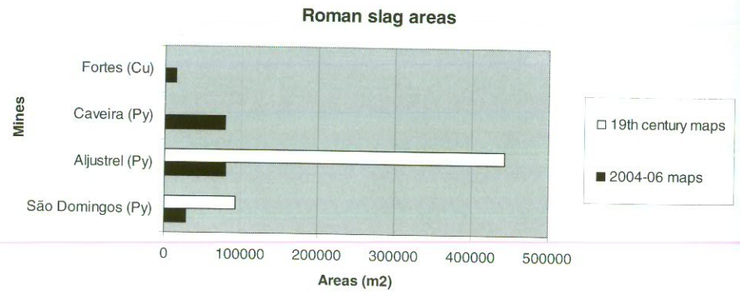

Fig. 4 – Global distribution of the Roman slag areas in IPB Portuguese sector.

8. Concluding remarks

During Roman times, several massive sulphide deposits in the Portuguese sector of the IPB were intensely exploited; up to a maximum known depth of 118 m at Algares, Aljustrel (the Roman *Vipasca*). Detailed geological and mining mapping of Caveira, Aljustrel and São Domingos (Cu, Pb, and Zn-bearing pyrite masses) and of Fortes (Cu-rich lodes) allowed a better understanding of the Roman slag distribution at each site and so, of the mining operation scale at each location. Local topography, available water sources or streams, and iron hat size and

location, conditioned the Roman mining operations. Long outcrops, due to the general sub-vertical character of the IPB Portuguese pyrite deposits (e.g. Aljustrel, S. Domingos and Chança) originated elongated Roman mining works in Portugal and led to Portuguese IPB Roman mines generally smaller than their Spanish counterparts. Only Aljustrel and São Domingos are similar in size to the Spanish mines (e.g. Río Tinto, Tharsis and La Zarza). Remnants of *in situ* Roman slag piles were identified, sometimes including pottery fragments; they are key-areas of mining heritage that should be preserved, because of their archaeological and cultural significance. Our knowledge of the Roman mining in the IPB is still incomplete. In situ slag deposits are crucial to evaluate the economic impact of south Portuguese mining in the context of the Roman Empire. Chança, Lousal and Montinho, although known to have been exploited in Roman times have no in situ or disturbed Roman slag dumps identified. As it is not conceivable that the ore was exported for smelting elsewhere, this must be taken to mean that the slag was destroyed or is hidden and thus, that the economic importance of the Roman mining industry in the *Lusitania* Province is still under evaluated.

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