

The copper archaeometallurgy at Monte Novo dos Albardeiros (Reguengos de Monsaraz, Évora)

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ABSTRACT

The site of Monte Novo dos Albardeiros is located near the village of Campinho (Reguengos de Monsaraz, Évora). A small tell situated next to the magazine of a farmstead was partially excavated, revealing several phases of occupation. The known phases of occupation are: a third millennium fortified farmstead; a funerary structure of tholos type, datable to the second half of the third millennium; and above the collapsed structure of the latter, two funerary deposits of the Early Bronze Age.

Metal artefacts were recovered from all of these phases, including traces of local copper metallurgy from the two earliest phases of occupation of the site.

Through the EDXRF method, the composition of two awls, an axe and an alène, all manufactured from copper, was analysed.

A brief technico-cultural study is also presented, stressing the «archaeometallurgic continuity» of the site and the question of arsenical copper.

Key-words: copper archaeometallurgy – copper artefacts – third millennium cal BC – Chalcolithic

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RESUMO

O Monte Novo dos Albardeiros situa-se perto da povoação do Campinho (Reguengos de Monsaraz, Évora). Junto ao hangar de máquinas da casa agrícola, existe um pequeno tell, que foi objecto de uma escavação parcial. Revelou diversas fases de uso, sendo as principais, por ordem decrescente de antiguidade, uma quinta fortificada da primeira metade do 3º milénio, uma estrutura funerária tipo tholos, datada da segunda metade do 3º milénio, e, sobre os derrubes de esta última, duas deposições funerárias da Antiga Idade do Bronze.

Nestes três conjuntos foram recolhidos artefactos metálicos, nos dois primeiros associados a traços de arqueometalurgia local do cobre.

Usando o método EDXRF, estuda-se a composição de dois furadores, um machado e uma alène, todos de cobre, provenientes de estes contextos.

Faz-se uma rápida integração técnico-cultural, sendo sublinhada a «continuidade arqueometalúrgica» do sítio e referida a questão do cobre arsenical.

Palavras-chave: arqueometalurgia do cobre — artefactos de cobre — 3º milénio a.n.e. — Calcolítico

1. INTRODUCTION

The visible part of the prehistoric site of Monte Novo dos Albardeiros consists of a small *tell*, situated near the village of Campinho (Reguengos de Monsaraz, Évora). Details of its location, discovery and general issues related to the site have already been published (Gonçalves, 1988-89, 1989 and 2005; Gonçalves & Alfarroba, *forthcoming*).

In an nut shell, the site was initially settled in the transition from the fourth to the third millennium cal BC. During the first half of the third millennium a fortified farmstead, of archaeometallurgists, was located at the site.

The area is wealthy in native copper and the collective burials of this phase are present in dozens of monuments built specifically for the purpose (passage graves – in Portuguese *antas* – and *tholoi*), or reused (earlier passage graves). During the second half of the third millennium a structure with a false dome, of *tholos* type, was built over the abandoned ruins of the monumental hollowed tower of the fortified farmstead. This structure had, from its earliest phase of occupation, a funerary function with remains of archaeometallurgy.

During a later phase two vases, found over a mass of decomposed human bone, were deposited over the ruined structure. Inside each of the vases was discovered an *alène*, a commonly found artefact during the European Early Bronze Age. This chronology is also confirmed by the form of the vases.

Although archaeological work at the site, directed by one of the authors (VSG), revealed the building and artefactual sequence of its occupation, continuity of this work is essential to understand if:

1. the different phases of occupation follow each other in rapid sequence, or are gaps in occupation;
2. the timeframe for the first occupational phase of the site equals the second or not;
3. there are other deposits dated to the Early Bronze Age.

In relation to the latter the proximity of a large Bronze Age necropolis of cist burials, located near Monte dos Cebolinhos (Gonçalves e Calado, 1990-91), may be important.

The absolute chronology for the site (^{14}C) indicates the following:

1. 2886-2460 cal BC, 2 σ (ICEN-530), for the fortified farmstead;
2. 2470-1910 cal BC, 2 σ (ICEN-529), for the *tholos* structure.

2. METALLIC ARTEFACTS

The artefacts forming the subject of the present study come from archaeological contexts dated to the first half of the third millennium (awl M.14-24), the second half of the third millennium (awl M.12-1 and axe K12.67+69), and the early centuries of the second millennium (*alène* J.11-97b).

The two burrowing artefacts, awls M.14-24 and M.12-1, correspond to different phases of use when they were abandoned, the first being well preserved, apparently without traces of intensive usage, while the second is bent but without damage to its distal end. Although corrosion makes the analysis of the proximal extremity of L.14-24 difficult, awl M.12-1 shows a spatulated finish similar to the proximal extremity of axe K12.67+69. Could this represent a multifunctional artefact, where both extremities were used?

Axe K12.67+69 is well preserved and seems to have been a votive object, since no traces of use are present on its distal extremity, usually in the form of blunting or damage to the cutting edge. The *alène* J.11-97b, seems undoubtedly to be a votive piece, found deposited in the interior of vase J.11-97, covered by the sediment that filled the latter until the moment of discovery.

Two other groups of artefacts, not available for analysis before the completion of this paper, consist of a large copper object of undefined use (axe ?), with a bent extremity, and another *alène* found in the same conditions as that in vase J.11-78 (image published in Gonçalves, 1988-89) – See, in this paper, Figure 2.

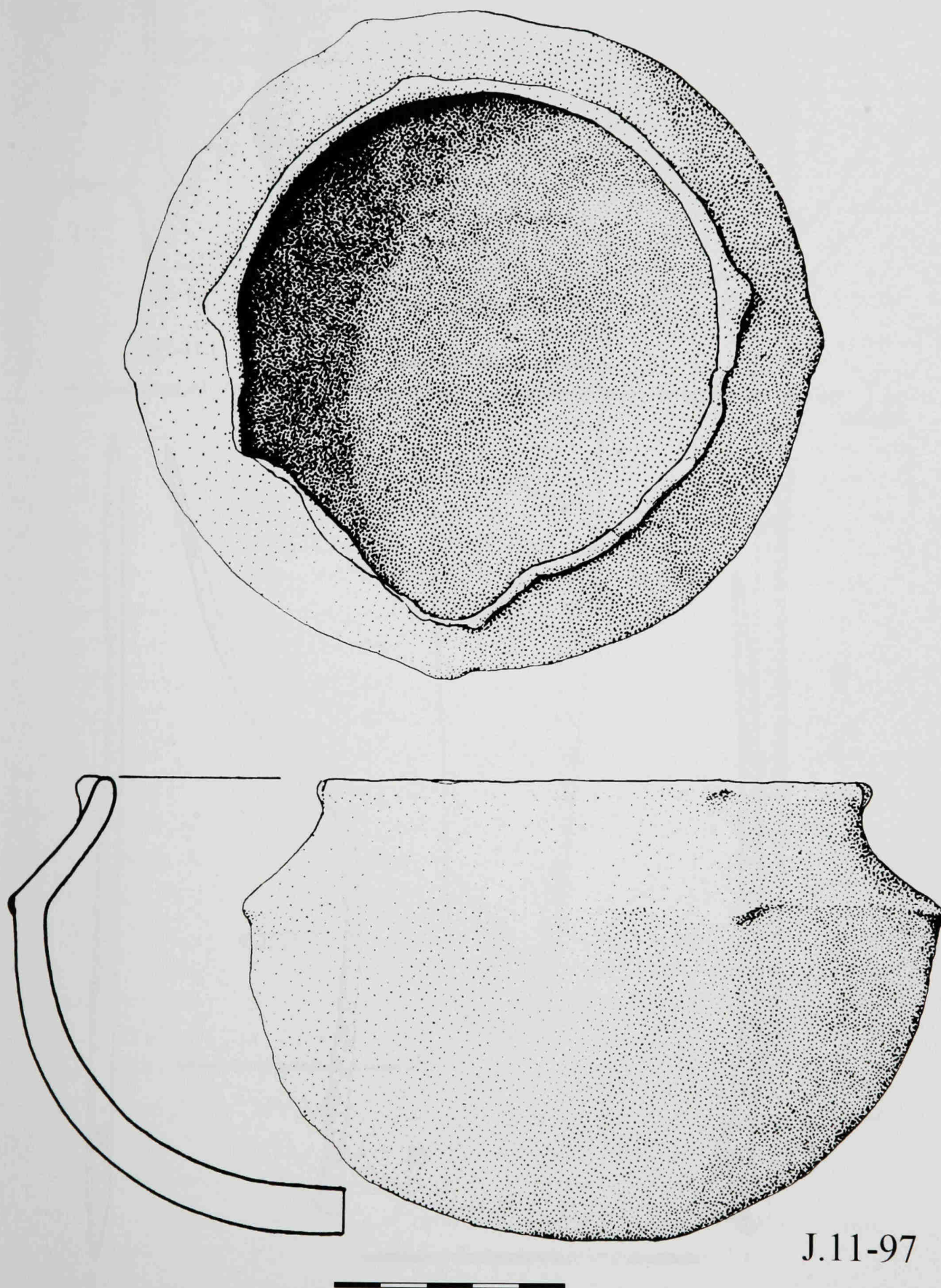


Fig. 1 – The copper *alène* J.11-97b, was found inside the Early Bronze Age carinated bowl, *apud* Gonçalves, 1988-89.

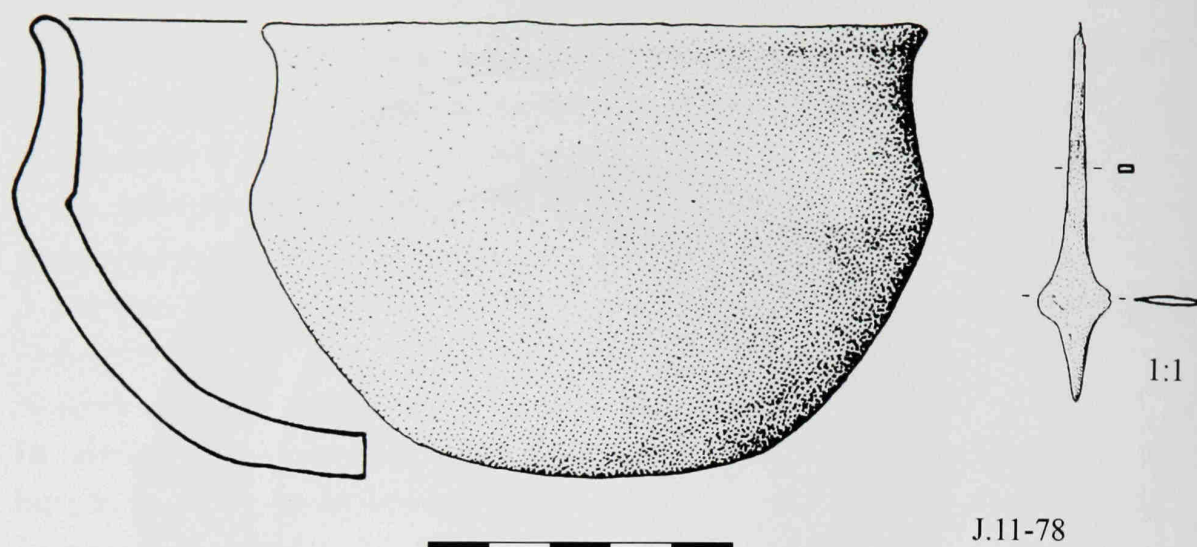


Fig. 2 – The Early Bronze Age soft carinated bowl, was found inside the copper *alène* J.11-78b, *apud* Gonçalves, 1988-89.

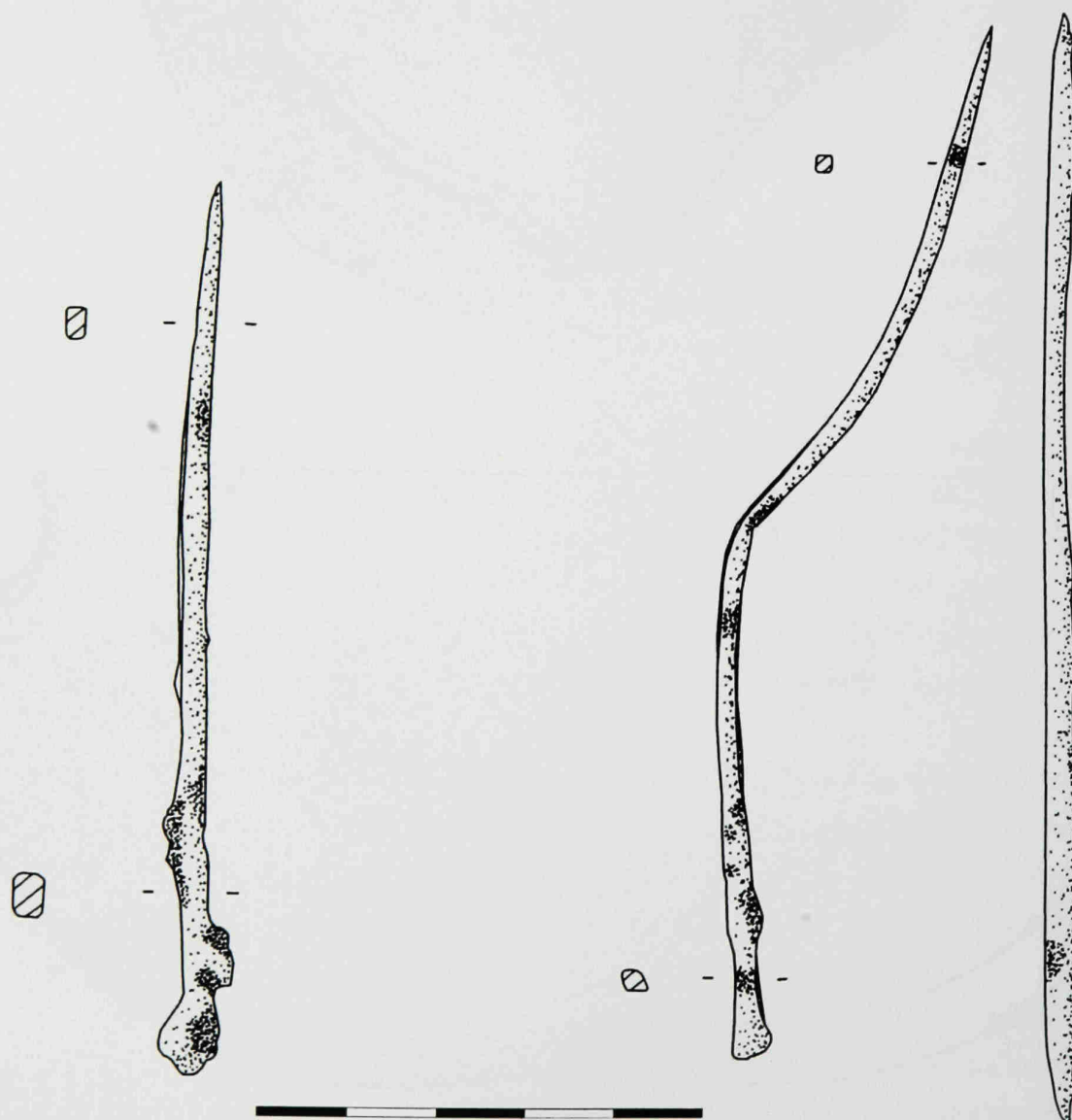


Fig. 3 – The awl M.14-24, from the first half of the 3rd millenium cal BC. Weight: 8,73 gr. H: 101,67 mm.Th: 4,05 mm.

Fig. 4 – The awl M.12-1, from the second half of the 3rd millenium cal BC. Weight: 6,35 gr. Th: 2,90 mm

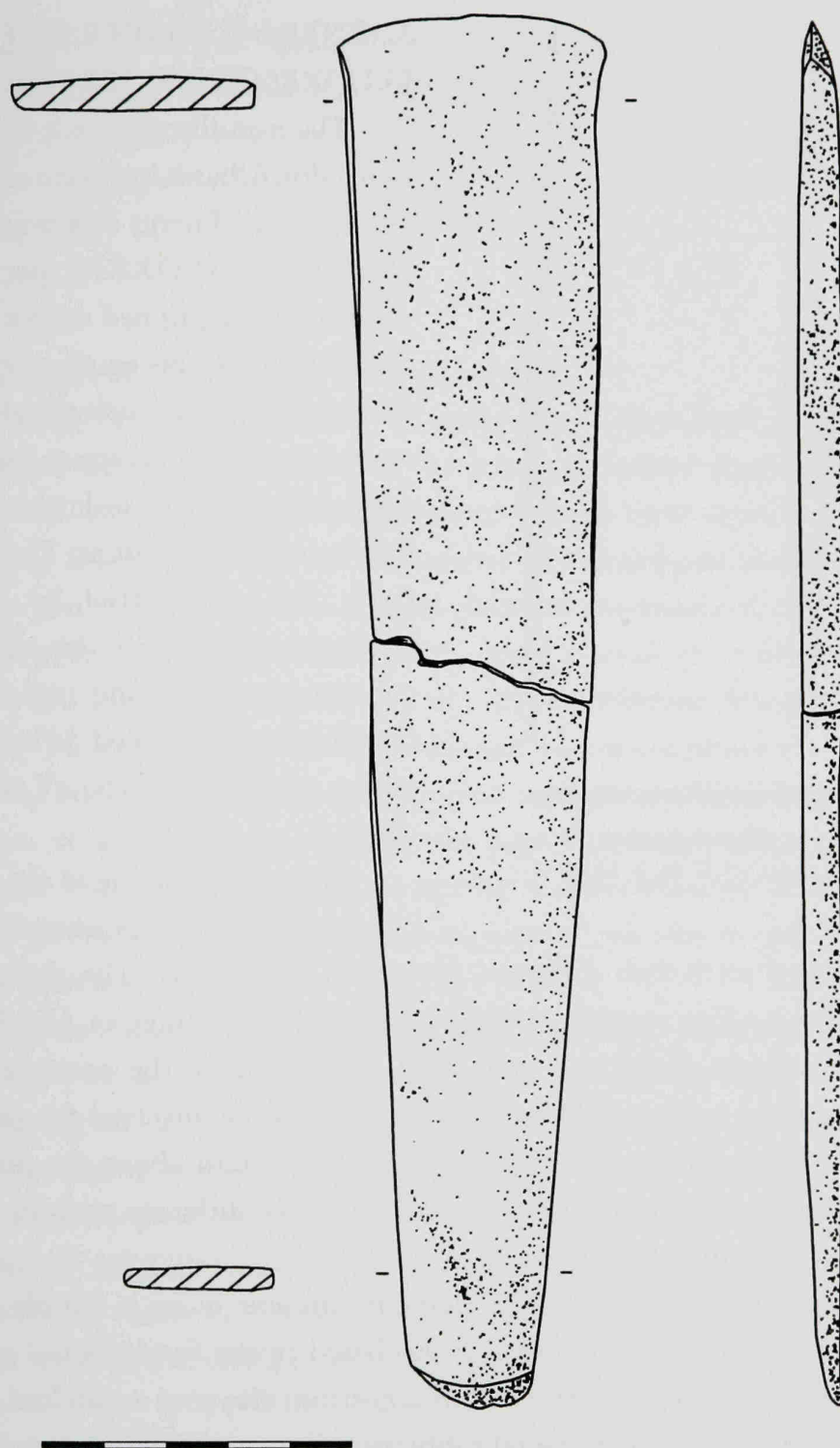


Fig. 5 – The axe K.12-67+69, from the second half of the 3rd millenium cal BC.
Weight: 357,72 gr. H: 22,7 cm

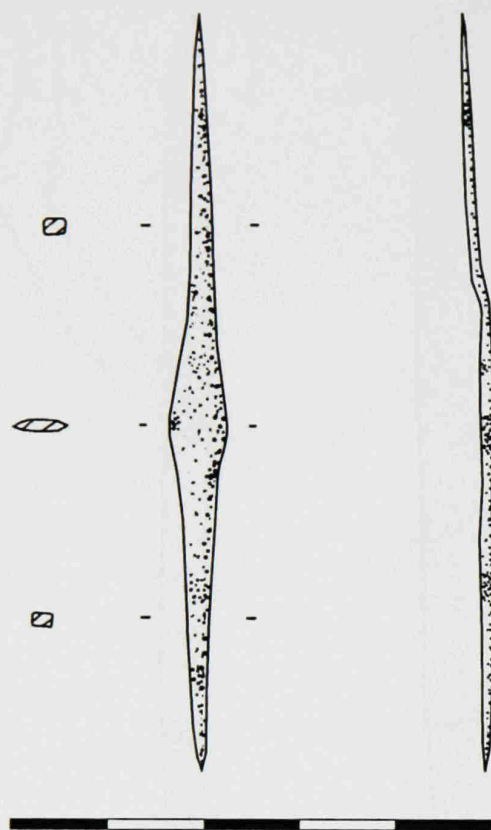


Fig. 6 – The *alène* J.11-97b, beginnings of the 2nd millennium cal BC. Weight: 1,57 gr. H: 78,35 mm. Th: 1,08 mm.

3. ENERGY DISPERSIVE X-RAY FLUORESCENCE SPECTROMETRY

The metallic materials from Monte Novo dos Albardeiros were analysed in a commercial Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometer – *KeveX 771*, equipped with a rhodium X-ray tube as the primary excitation source. The system includes also several secondary excitation targets and suitable radiation filters in order to optimize the excitation conditions. The characteristic X-rays emitted by chemical elements present in the sample are collimated at 90 ° and measured in a liquid nitrogen cooled Si(Li) detector with a resolution of 165 eV at 6,4 keV (Fe- K_{α}).

The determination of the chemical composition of copper-based artefacts

is accomplished with two different excitation conditions. The characteristic radiation from a silver secondary target, with a high voltage of 35 kV and an intensity of current of 0.5 mA, was used to investigate the presence of iron, nickel, zinc, copper, arsenic and lead. The second condition involved the gadolinium secondary target, with 57 kV and 1.0 mA, to ascertain about the presence of tin and antimony. The artefacts were analysed in two different surfaces and with a real measuring time of 300 s.

The quantification of chemical elements present in each sample used the EXACT computer program (Kevex, 1990) based in the fundamental parameter method (Tertian and Claisse, 1982). This algorithm also uses a standard reference material to establish the experimental calibration parameters. These were calculated with the analysis of one reference material (Phosphor Bronze 551 from British Chemical Standards) comparable to the studied artefacts. The detailed description of the overall experimental procedure was previously published (Araújo *et al.*, 1993).

The accuracy of the method was determined with the quantification of two reference materials (Phosphor Bronze 552 from British Chemical Standards and Bronze 5 from Des Industries de la Fonderie). The calculated errors

regarding the elemental quantification were as follows: 1% for copper, 7% for arsenic and 10% for iron. The quantification limits (Currie, 1968) for elements usually present in the studied alloys were: Fe – 0.03 %, Ni – 0.05 %, Zn – 0.03 %, As – 0.10 %, Sn – 0.02 %, Sb – 0.03 % and Pb – 0.04 %.

3.1. Limitations of EDXRF in archaeometallurgy

The weak X-ray penetration in materials with a high atomic number (Grieken and Markowicz, 1993), as the present studied copper-based alloys, results in the quantification of a superficial sample layer usually with a depth inferior to a few hundred micra. In general archaeological metallic materials present an external layer with an altered chemical composition, due to corrosion processes which took place during the long period of artefact deposition (Walker, 1980 and Balasubramaniam *et al.*, 2004). Different elemental stabilities, as well as, dissimilar stabilities of the formed corrosion products, produce the preferential enrichment of certain elements in the superficial alteration layer. In the case of arsenical copper alloys the superficial enrichment in arsenic is well-known. Furthermore, the iron content is also enhanced in the alteration layer due to the incorporation of soil elements and also to the formation of iron rich corrosion products.

Consequently, this heterogeneous superficial layer should be removed so that the analysed section can be representative of bulk material. Unfortunately, when studying archaeological artefacts it is neither possible nor desirable to use superficial cleaning methods, since those procedures can damage or modify the valuable archaeological materials.

Although, EDXRF accurate results are heavily dependent on the homogeneity of the analysed material, its non-invasive characteristic turns it in the most important analytical technique in the study of archaeological and museological artefacts.

In our study a gentle mechanical cleaning (e.g. water and scalpel cleansing) was applied to the surface of each artefact in order to minimize the above mentioned problems. The artefacts were analysed before and after the application of this method to ascertain its efficiency. The obtained results demonstrated different cleaning outcomes for each artefact surface, however in all instances the reduction of the iron content was the main observed variation (Fig. 1), undoubtedly as the result of the partial removal of soil-based products.

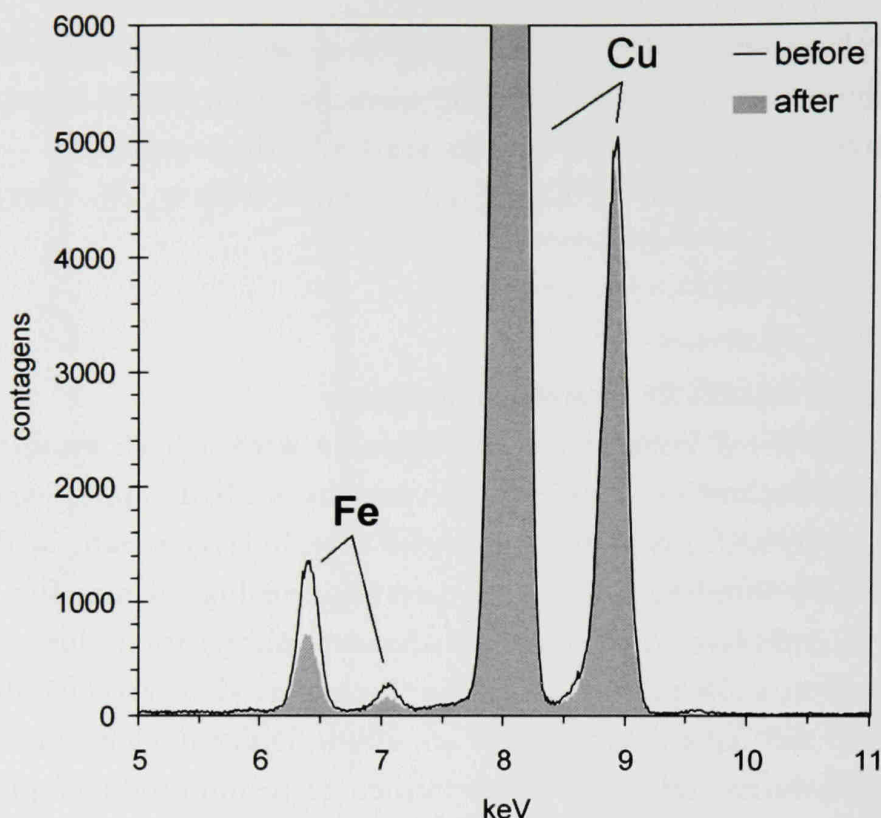


Fig. 7 – EDXRF spectra of awl (MNAL M.14-24) presenting the reduction of the iron content after the gentle cleaning procedure.

The following discussed results concerning the metallic materials from Monte Novo dos Albardeiros were obtained after the cleaning method and are interpreted considering the above mentioned EDXRF spectrometry limitations in archaeometallurgy.

4. RESULTS

The non invasive analysis of metallic artefacts recovered in the archaeological field work at Monte Novo dos Albardeiros are presented in table 1.

Table 1 – Chemical composition of metallic artefacts from Monte Novo dos Albardeiros (values in %; n.d. – not detected)

Artefact	Cu	As	Fe
Awl	97.9	n.d.	1.8
(MNAL M.14-24)	98.0	n.d.	1.7
Awl	99.4	<0.10	0.2
(MNAL M.12-1)	99.4	<0.10	0.3
Axe (fragment 1)	97.4	2.3	0.3
(MNAL.K.12-67)	97.3	2.5	0.2
Axe (fragment 2)	96.9	3.0	<0.03
(MNAL.K.12-69)	96.7	3.2	<0.03
Alène	94.9	4.8	0.09
(MNA. J.11-97b)	93.8	5.8	0.06

Apparently, two different groups of artefacts can be distinguished accordingly to their composition. The two awls (MNAL M.4-24 and MNAL M.12-1) are constituted by relatively pure copper matrix with traces of arsenic in the second artefact. The obtained iron contents, as mentioned before, are influenced by soil contamination, which was not totally removed by the applied cleaning method. The absence of other impurities and the low arsenic contents point out to the use of copper rich ores which can be easily reduced, as the copper carbonates mined in the Barrancos Mining Area. This mining area is located approximately 40 km from the settlement and was also a pre-historic source of native copper (Cardoso *et al.*, 2002). The Monte Novo dos Albardeiros is located in the Faixa de Arraiolos – Stº Aleixo (Martins and Borralho, 1998), an area with several copper mineralizations (e.g. Reguengos), as well as ancient copper mines (e.g. Monte do Trigo), some of them worked in prehistoric times – Rui Gomes and Monte do Judeu (Cardoso *et al.*, 2002).

The second group includes the two fragments of the same axe (MNAL.K12.67 and MNAL.K12.69) and the *alène* (MNAL J.11-97-b). A copper matrix with significant arsenic contents constitutes these. Regarding the recognized enrichment of this element in the superficial corrosion layer, it is difficult to ascertain about the real content and origin of the arsenic in the studied artefacts – it could be a result of the use of arsenic rich copper ores or it could be an addition to the copper source.

Marechal (1985) compared the arsenical contents with other minor and trace elemental contents in order to establish the copper ores used in the artefact production, concluding that only contents higher than 4 % are indicative of deliberate additions. Regarding the production of arsenical copper alloys, it should be mentioned that, accordingly to Ortiz (2003), artefacts with low arsenical content (< 2 %) can not be further toughened by hammering. It should be also considered that a very high arsenic content produces a rather hard alloy which is very difficult to work.

Then it appears that the relatively low arsenic content obtained in the axe [2.4 % – 3.1 %], should point out to the natural presence of this element in the copper alloy, probably resulting from the use of an arsenic rich copper ore. Despite being a highly volatile element, arsenic also presents a reduced affinity to the slag formed during the smelting process and within certain reducing conditions is preferentially associated with the metallic copper during this metallurgical operation (Ortiz, 2003). The higher arsenical content in the *alène* [4.8 % – 5.8 %] could indicate a planned addition but the alloy could as well be the result of the above mentioned arsenic rich copper sources.

The low iron concentration of the *alène* [0.06 % – 0.09 %] and, in particular, of the axe (fragment II) [<0.03 %], point out to the fairly reducing environment of the smelting operation. The absence of a good reducing environment prevents the reducing of the iron minerals present as impurities in the copper ore and therefore the metallic copper formed has low iron content (Craddock, 1995). This primitive smelting process was conducted in simple open ceramic vase heated from above (Rovira and Ambert, 2002). The process is specially adapted to copper carbonates, ores which are easy to process and were commonly used in this region of the Iberian Peninsula (Rovira, 2002). In the south area of the Portuguese territory there are several examples of the use of this primitive smelting operation – Castelo Velho de Safara (Moura), Porto Mourão (Moura), São Brás 1 (Serpa) and Três Moinhos (Beja) (Soares *et al.*, 1996).

5. ARCHAEOMETALLURGICAL INTEGRATION

In the South West of the Iberian Peninsula, artefacts constituted by arsenical copper alloys are present since the establishment of metallurgy, while the pure coppers are relatively rare during the Copper Age (Ortiz, 2003). The presence of this element in the copper matrix seems to derive from its occurrence, with variable concentrations, in the oxide and carbonate copper ores. However, the control of the final arsenical content should be quite difficult to achieve and there are several ways that probably would give some results: i) intentional selection of ores with different arsenical content; ii) induced arsenical losses during the foundry operations; iii) induced arsenical losses during cold working in oxidizing conditions. Therefore, it seems that the majority of the arsenical copper alloys are the result of its natural presence in the copper ores used, fact that could explain the lack of relation between the artefact types and the arsenical content (Ortiz, 2003).

The analysis of artefacts from some sites in the Portuguese territory, namely Liceia (Oeiras) and Outeiro de São Bernardo (Moura) point out to the same conclusions. Materials from Liceia present a continuous arsenical content variation [0 % – 5.5 %] which was interpreted as the direct result from the copper ores used (Cardoso *et al.*, 2002). In the latter settlement, it was considered that despite the limitations of the non invasive analysis, the pattern seem to be similar to the materials from the first site and so the arsenical contents seem to derive from the copper sources (Cardoso *et al.*, 2002).

The study of metallic artefacts from different settlements located in the middle Guadiana River (Soares *et al.*, 1994) seem to reveal a different reality –

arsenical copper alloys only become significant during the Late Copper Age and before this period the metallic artefacts are composed by pure copper, obtained first from native copper and latter from copper oxide and carbonate ores. The latter arsenical copper alloys were probably produced by the addition of arsenic ore to the copper smelting charge, using the co-reduction process (Lechtman and Klein, 1999). The occurrence of a fragment of pyrite ore associated with arsenopyrite in the Late Copper Age occupation of Castelo Velho de Safara (Moura) supports this proposal (Soares *et al.*, 1985). Furthermore, considering the studied sites in this area, it seems that, contrary to the Spanish side of the South West Iberian Peninsula, the pure copper artefacts are not so uncommon. From a total of 49 analysed artefacts, about 10 % are composed by pure copper and approximately 30 % have a low arsenical content (< 1 %).

The results of recent work (Valério *et al.*, forthcoming) concerning the study of metallic artefacts and ores from the Campaniform occupation of Porto das Carretas (Mourão) also point out to a similar conclusion. The artefacts are composed by copper with significant arsenical contents, contrary to the ore fragments which are free from any traces of arsenic and therefore couldn't originate the above mentioned alloys without the deliberated addition of an arsenical source.

Despite the reduced number of analysed artefacts the new results from Monte Novo dos Albardeiros seem to support this latter thought regarding the not yet clear metallurgical pattern of the Copper and First Bronze Ages in the middle Guadiana River area. The awl (MNAL M.14-24) from the previous site occupation (2886-2460 cal BC 2s) didn't present any traces of arsenic. The second awl (MNAL M.12-1), from a second occupation period (2470-1910 cal BC 2s), shows some traces of arsenic. The axe (MNA K.12-67 and MNAL K.12-69), belonging also to this second occupation stage, already presents a significant content of arsenic, while the alène (MNAL.1(86) L.12), which belongs to the First Bronze Age, present an even higher arsenical content.

The contradiction between the metallurgic evolution that took place in this region and the one studied in the Spanish side of the South West Iberian Peninsula could probably be explained by the existence of numerous copper ores (oxides and carbonates) free from arsenic and from which it should be rather straightforward to obtain a pure copper using rather primitive metallurgical operations.

6. DISCUSSION

Based on these results it could be concluded that in Monte Novo dos Albardeiros the metal is present since the first half of the third millennium BC, while the arsenical coppers

only occur during the second half of the third millennium cal BC – beginnings of the next millennium. The First Bronze Age continues the metallurgical tradition from the previous period, i.e. the copper tin alloys are still absent, as in the other settlements from the region, such as Cerros Verdes 3, with an arsenical copper arrow (Araújo and Valério, 2005) belonging to the Early Bronze Age of the Southern Iberian Peninsula.

The archaeometallurgy of copper was profoundly connected with the process of transformation of the economy, society, and even the magico-religious subsystem, brought about by the *Secondary Products Revolution* in the Centre and South of Portugal, from the beginning of the first half of the third millennium cal BC (Sherratt, 1981; Gonçalves, 1989, 1993).

The great changes in the utilisation of the territory are concentrated in the adoption of an intensive agriculture, until then unknown, aided by the use of the plough and wagon, facilitating the transport of the crop of grasses, and the integration of archaeometallurgy as a local activity.

This archaeometallurgic activity meets the local needs of those who practised it, while overproduction, in the form of ingots or finished objects, was traded to places without native copper resources. Small copper ingots could be easily transported over considerable long distances, to fortified settlements like Vila Nova de S. Pedro, Liceia, Zambujal, Moita do Ladra, among others.

Small settlements such Cabeço do Pé da Erra, Coruche, also revealed archaeometallurgical practices (Gonçalves, 1982, 1983-84). Although the site is small, and relatively isolated (in the third millennium cal BC, a small island), and the scarcity of artefacts, metallurgy was practised, probably in a great hearth found close to the dwellings, where a schist plaque workshop was also found.

The site of Monte Novo dos Albardeiros is a fortified farmstead strategically located on the schist–granite fold, with its back turned to the first, depopulated and economically insignificant, to control the rich territory of the latter, where soil use capacity is rated of categories B and C (also A along narrow stretches), and with native copper.

The vertical and long time span distribution of copper artefacts at the settlement of Monte Novo dos Albardeiros demonstrates their presence for a thousand years from the beginning of the third millennium, showing remarkable continuity confirmed in the area, in the context of the ritual of death, by the construction and collective use of *tholoi* and the reutilisation of older megalithic (orthostatic) monuments (as in STAM-3 or Anta Grande do Olival da Pega, see Gonçalves, 1999, 2003).

The presence of symbolic ceramics (Gonçalves, 1988-89) and two figurines of the well known Goddess of the South of the Iberian Peninsula (Gonçalves, 2005), relate the magico-religious system.

The question of arsenical copper remains unanswered and only the analysis of many more copper artefacts will convey a better and reliable knowledge than at present.

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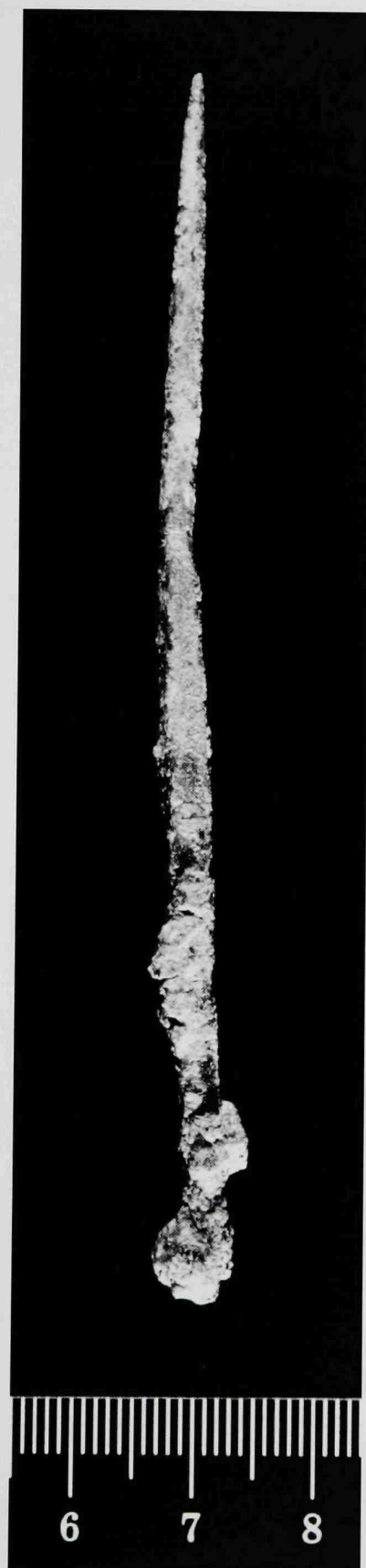
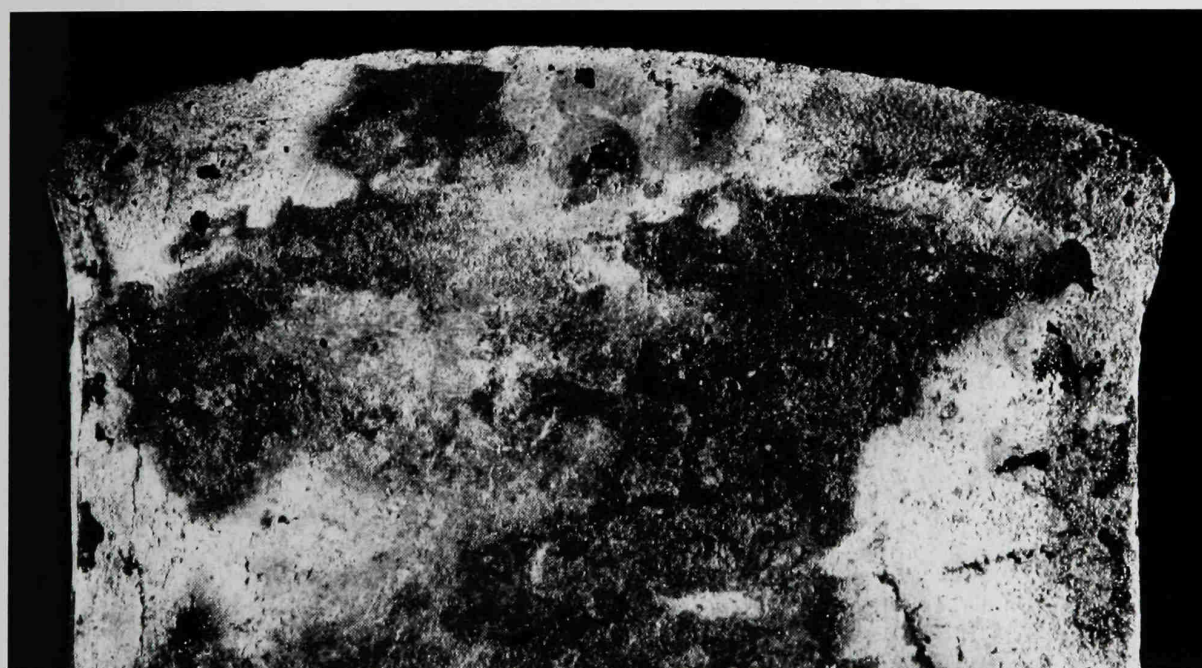
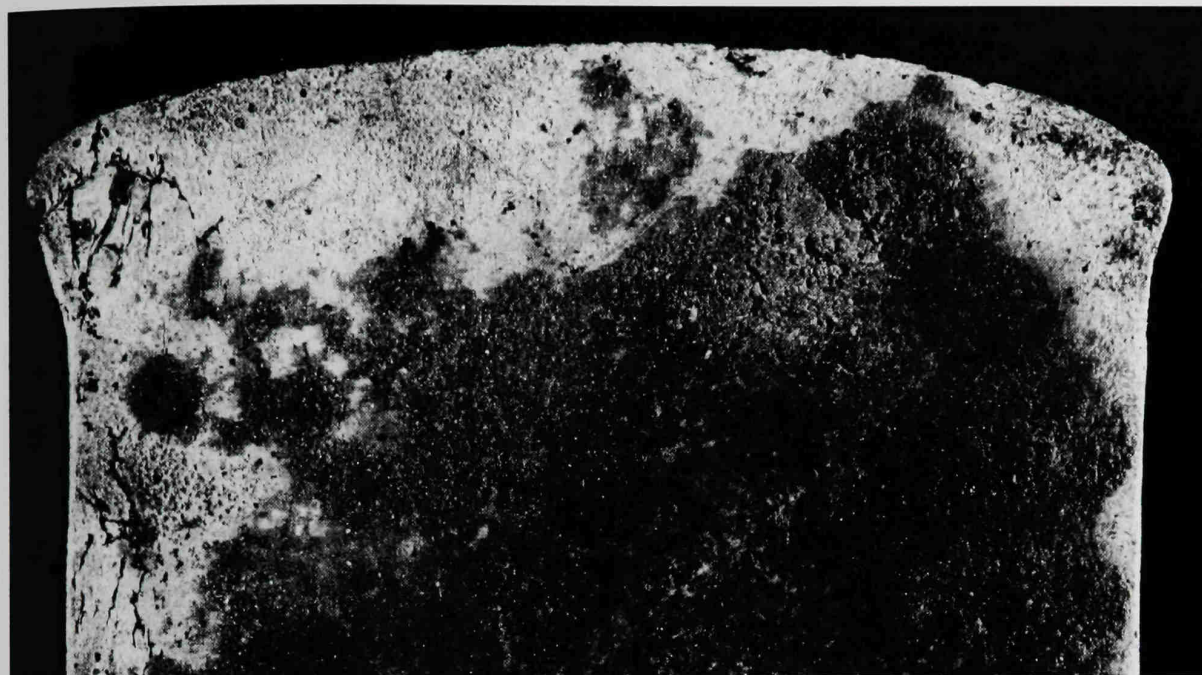


Fig. 8 – The awl M.14-24.



Fig. 9 – Two sights of the axe K.12-67+69 (front and rear).



Figs 10 e 11 – Two sights of the cutting edge of the axe K.12-67+69 (front and rear view).



Fig. 12 – The awl M.14-24.



Fig. 13 – Two sights of the distal e proximal extremities of the awl M.14-24.

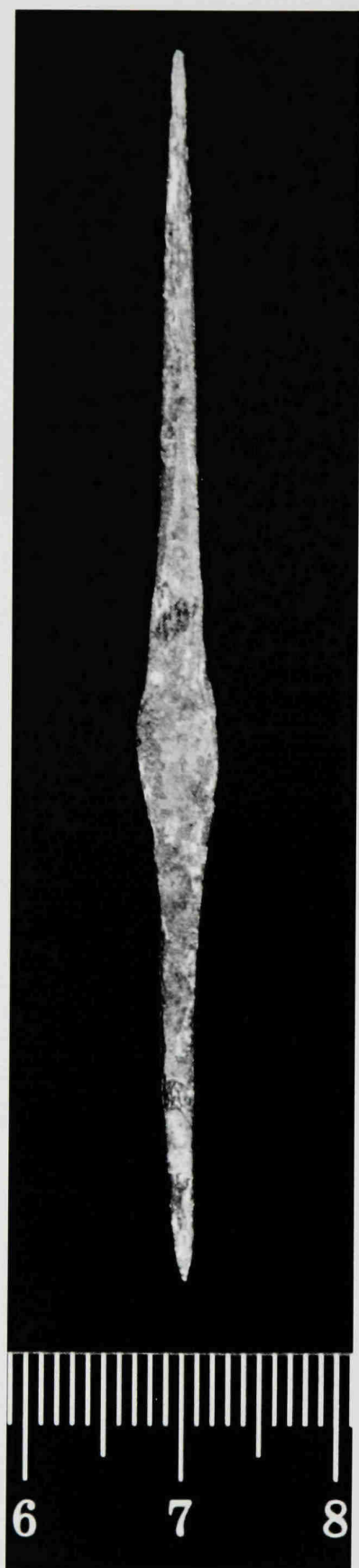


Fig. 14 – The *alène* J.11-97b.



Fig. 15 – The Early Bronze Age carinated bowl J.11-97b, within of which was found the copper *alène*, front view.

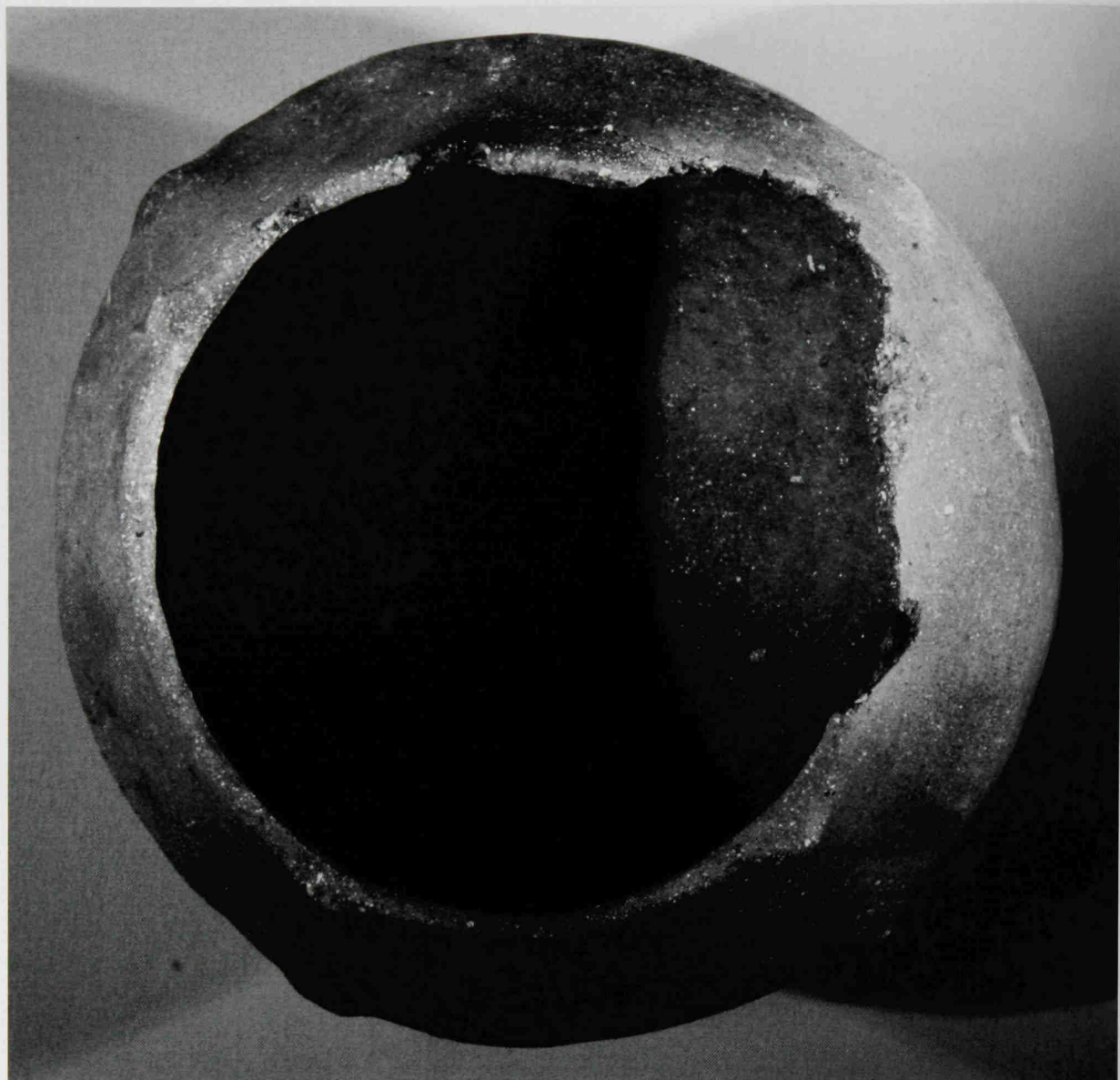


Fig. 16 – J.11-97b, upper view.

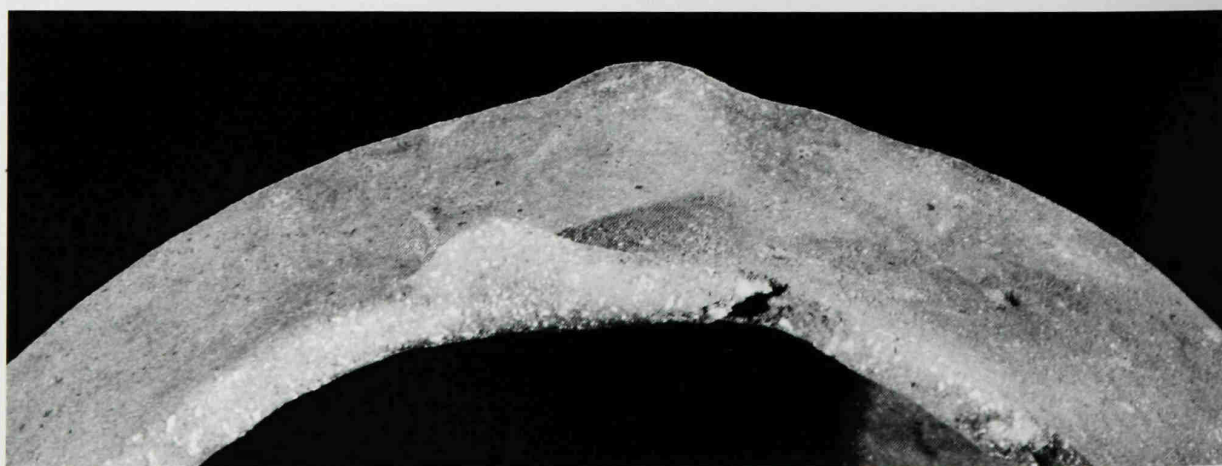


Fig. 17 – J.11-97b, upper view, detail.



Fig. 18 – J.11-97b, front view, detail