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The sanctuary of Hercules in Sesklo Region, Volos, Greece: an archaeometric approach of the archaic bronze objects

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ABSTRACT
The study of the bronze offerings obtained from the Sanctuary of Hercules in the area of Sesklo, Municipality of Volos, Thessaly, Greece is presented in this paper. The objects were examined initially with non-destructive followed with invasive methods in order to better understand their manufacture technology. The provenance of copper is also briefly discussed.

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1. Introduction

1.1. Sanctuary of Hercules

The Sanctuary of Hercules (Stamelou and Doulgeri-Intzesiloglou 2010) is located on the northern slopes of the hill named "Spartias" in the area of Sesklo, Municipality of Volos, Thessaly, Greece on the course of the ancient road that connected the ancient city of Pherae with its seaport, Pagasae (Figure 1). The Sanctuary was identified by a votive inscription on a bronze omphalos phiale (Figure 2) and it is the only confirmed excavated site of Hercules worship, outside the city, however, within the region of Pherae (Figure 3). The cult of Hercules in this Sanctuary dates back to the Archaic period (sixth century BC), when Thessaly was organized into four main regions ("tetrades") and big cities, such as Pherae, consolidate their power. The Sanctuary is still present during the Hellenistic period (second century BC).

During the excavation works, part of the stone foundation of a rectangular structure came to light, which was interpreted as an altar, measuring about 3.00 m × 4.00 m and dating to the second century BC. Around the foundation, a dense layer of stone piles, covering deposition pits was unearthed, from which many artefacts dated to the second century BC, including numerous iron and bronze offerings, were collected.

Copper-based artefacts were produced for several everyday purposes and among them to fulfill the people's sacred offers either to the dead or to their gods. To find objects from different chronological periods is a common phenomenon in ancient sanctuaries, as offerings of worshippers were packed over the years and the people in charge were forced to remove them. Most often, they were thrown into wells, rivers or in specially dug pits, as in the case of the Sanctuary of Hercules. Of course, all these offerings remained sacred, and therefore the Hellenistic construction (interpreted as an altar) was founded above them.

Taking the chance of the considerable amount of copper alloy objects revealed from the Sanctuary of Hercules, this study was undertaken in order to understand the manufacturing technology of the objects, to determine the metal or the alloys used, to point any imported artefacts and finally to determine the possible copper ore sources.

2. Methods

For the purpose of this study, 27 copper-based votive objects have been selected for archaeometric analysis. For the analysis of the objects, both invasive and non-invasive methods were used.

2.1. Analysis in the laboratory – portable X-ray fluorescence

The majority of the samples were subjected to X-ray fluorescence (pXRF) analyses which were performed at the Conservation Laboratories of the Ephorate of Antiquities in Volos.

The pXRF used is a portable, though not handheld, ED-XRF spectrometer developed at the Institute of Nuclear Physics, NCSR Demokritos.
The XRF spectrometer consists of an Rh-anode side-window, low power X-ray tube (50 W, 40 kV, 125 μm Be window), a PIN X-ray detector and a multi-channel analyzer (MCA) card. The analytical range of this portable XRF spectrometer extends from \( Z=14 \) (silicon) up to \( Z=92 \) (uranium). The device can operate under two distinct conditions: one unfiltered mode with the voltage set at 15 kV and a filtered one with the voltage set at 40 kV. Two laser pointers are mounted in the spectrometer head in such a way that the intersection point of their beams coincides with the crosspoint of the incident X-ray beam axis and the detector axis. The beam spot at the sample position has a diameter of less than 2

Figure 1. Map showing the location of the Sanctuary of Hercules on the course of the ancient road that connected the ancient city of Pherae with its seaport, Pagasae and its relation to other ancient cities.

Figure 2. Sketch of the bronze omphalos phiale (BE45972) with the votive inscription (drawing: E. Rini).
mm. The spectrometer head is attached in an X-Y-Z position, allowing its easy movement in the X-Y directions. (Karydas et al. 2009: 813-814)

The pXRF measurements were taken both on a clean metal and the patina. With the term “clean metal”, we mean the substrate metal layer on a cleaned surface free of obvious corrosion products. For each analysis, two spot measurements of 2 mm were taken in different areas in order to have more representative chemical compositions; it is known that many changes occur

Figure 3. The rectangular stone foundation of the altar, view from the West.

Figure 4. Phiale BE46153. Photograph through OM, magn 20x plus. Grain boundaries due to hammering, cuprite and paratacamite crystals.
**Figure 5.** Phiale BE46146. Photograph through OM, magn 50×. Copper sulphide inclusions are clearly visible throughout the body of the metal.

**Figure 6.** Phiale BE46161. Photograph through OM, magn 20×. Strain lines due to intensive hammering are obvious.
Figure 7. Phiale BE46152. Photograph through OM, magn 10×. Original surface measurement, 448 mm.

Figure 8. Phiale BE46159. Photomicrograph through EPMA, magn 400×. Severe cracking is obvious.
on the surface of the objects during their burial, and in this way one can test if the surface analysis is representative of the metal composition, as usually the substrate is also corroded (Scott 1985, 1991, 2002; Mezzi et al. 2012, 953).

The measurement parameters were time 300 s, in 40 kV, 30 μΑ. Prior to the analysis, the objects’ Certificate Reference Materials (BCR-691) were analysed to test the stability of the instrument.

2.2. Polarized light microscopy

Selected samples were mounted in resin blocks and polished in order to provide clean cross-section for analysis. Optical microscopy (OM) was used in order to examine the manufacturing techniques and shaping (Scott 2011), as well as any alteration occurred due to corrosion products (Tylecote 1979; Scott 1991). The OM was performed at the Research Science Laboratories of UCL Qatar.

2.3. Electron probe microanalysis–wave dispersive spectroscopy

Quantitative analyses were conducted with an EPMA–WDS (JXA-8100 Electron Probe Microanalyzer) at the Wolfson Science Laboratories of the Institute of Archaeology, UCL. Pure elements were used to calibrate the instrument as well as certified reference materials, namely brass 42.23.2 and leaded bronze 50.04.4 (Bureau of Analysed Samples Ltd). The instrument worked with an accelerating voltage of 20 kV, spot size set to 0 μm, magnification ×1000 and beam current of 50 nA. The data presented here for each sample are the average of the individual measurements.

3. Results and discussion

Both macroscopical and microscopical examination revealed that 24 out of 27 objects are hammered and annealed (Figure 4), that is, the phialae, the ankle guards, the sheets, the vessels, the rings, the pins, the round object, the small shield and the omphalion. Sulphide slag inclusions are present in some of them (Figure 5) as well as strain lines showing the degree of hammering (Figure 6). Although some objects are extensively corroded, it was possible to determine their original surface and measure it (Figures 7–9).

During quantitative analyses with both pXRF (Table 1) and EPMA (Table 2), a set of nine elements were analysed, namely Fe, Co, Ni, Cu, Zn, As, Pb, Sn and Sb. In the cases that the patina was analysed with pXRF, Ca, K and Cl were also considered.

The analyses with pXRF presented in Table 1 were performed both on the surface metal and the patina.
It would be a mistake to compare the results of the two techniques as pXRF measurements are conducted on the substrate, with all the possible element enrichments that might have happened, while the EPMA was conducted in the metal core. Zinc (Zn) that was detected with pXRF in a monotonous presence of 0.3–0.4% w/t is absent in EPMA results and the same occurs with cobalt (Co) in some samples. As pXRF is an air path technique and zinc and cobalt are present in these monotonous values and absent in EPMA results, we attribute this to a malingering of the instrument. (Orfanou and Rehren 2014).

The quantitative examination of the samples showed that copper–tin alloy was used for the manufacture of the artefacts in a percentage of 4–11 wt-% tin. In the cases that tin is calculated in higher levels, with pXRF, this is attributed to its enrichment on the surface due to corrosion.

Lead has been broadly used during the antiquity in copper alloys. As it is known by the literature (Giumlia-Mair 2005), its addition improves the fluidity and the castability of the alloy. Only three objects seem to be cast, namely the shaft, the handle of the urn and the arrowhead. In the rest of the objects, either lead is totally absent or it is present in very small amounts.

| Table 1. Quantitative results of the chemical analyses performed with pXRF both on surface metal and the patina. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| pXRF | Sample ID | Area | Ca | Fe | Co | Ni | Cu | Zn | As | Pb | Sn | Sb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Phialae | BE 46160 | Surface metal | nd | 1 | 0.2 | nd | 92 | 0.2 | 0.5 | nd | 6 | nd |
| Patina | 3 | 2 | nd | nd | nd | 80 | 0.5 | nd | 14 | nd |
| BE 46159 | Surface metal | nd | 0.1 | nd | 93 | 6 | 0.1 | nd | 7 | nd |
| Patina | 11 | 3 | nd | nd | nd | 68 | 0.1 | nd | 13 | nd |
| BE 46136 | Surface metal | nd | 1 | 0.7 | 0.3 | 93 | 0.3 | 0.2 | nd | 5 | 0.1 |
| Patina | nd | 1 | nd | 93 | 0.4 | nd | 17 | nd |
| BE 46161 | Surface metal | nd | nd | nd | 0.1 | 93 | 0.3 | 0.1 | nd | 18 | nd |
| Patina | 7 | 1 | nd | 72 | 0.1 | nd | 18 | nd |
| BE 46135 | Surface metal | nd | 1 | nd | 93 | 1 | nd | 5 | nd |
| Patina | 9 | 1 | nd | 72 | 0.4 | nd | 13 | nd |
| BE 46137 | Surface metal | nd | 0.5 | nd | 93 | 0.3 | nd | 0.3 | 6 | nd |
| Patina | nd | 1 | nd | 84 | 0.1 | nd | 15 | nd |
| With inscription | BE 45972 | Surface metal | nd | 1 | nd | 92 | nd | nd | 7 | nd |
| Patina | nd | 1 | nd | 83 | nd | nd | 15 | nd |
| Omphalion | BE 46132 | Surface metal | nd | 1 | 0.1 | nd | 89 | 1 | nd | 0.1 | 8 | nd |
| Patina | nd | 1 | nd | 80 | 0.1 | nd | 18 | nd |
| Handle | BE 46144 | Surface metal | nd | 1 | nd | 79 | 6 | 2 | 11 | 0.4 |
| Patina | 14 | 1 | nd | 61 | nd | 6 | 21 | nd |
| Sheet | BE 46145 | Surface metal | nd | nd | nd | 87 | 2 | nd | 8 | nd |
| Patina | nd | 2 | nd | Nd | 53 | nd | nd | 45 | nd |
| Ankle guard | BE 45969 | Surface metal | nd | nd | nd | 88 | 0.3 | nd | 11 | nd |
| Patina | nd | 2 | nd | 74 | 0.1 | nd | 19 | nd |
| Ankle guard | BE 45970 | Surface metal | nd | nd | nd | 88 | 0.3 | nd | 11 | nd |
| Patina | nd | 2 | nd | 74 | 0.1 | nd | 19 | nd |
| Pin | BE 45960 | Surface metal | nd | 1 | 0.2 | 91 | 0.3 | 2 | 5 | nd |
| Patina | 12 | 4 | nd | 53 | 0.4 | 2 | 29 | nd |
| Pin | BE 46128 | Surface metal | 1 | 0.4 | nd | 89 | 0.2 | nd | 6 | nd |
| Patina | 6 | 2 | nd | 73 | 2 | 2 | 15 | nd |
| Double pin | BE 46123 | Surface metal | nd | 0.3 | 0.1 | 94 | 0.2 | 0.1 | 5 | 5 | nd |
| Patina | 14 | 2 | 0.1 | 65 | nd | 0.2 | 18 | nd |
| Ring | BE 46125 | Surface metal | nd | 0.1 | nd | 92 | 1 | nd | 6 | nd |
| Patina | nd | 2 | 0.1 | nd | 80 | 2 | nd | 15 | nd |
| Round object | BE 46129 | Surface metal | nd | 1 | 0.2 | 87 | 0.1 | nd | 11 | nd |
| Patina | nd | 1 | nd | 77 | 0.1 | nd | 5 | nd |
| Arrow head | BE 46130 | Surface metal | nd | 1 | 0.1 | 85 | 1 | 4 | 8 | nd |
| Patina | nd | 2 | nd | 59 | 1 | 4 | 34 | nd |
| Shaft | BE 46126 | Surface metal | nd | 1 | nd | 89 | 1 | nd | 6 | 3 | nd |
| Patina | nd | 2 | nd | 81 | nd | nd | 10 | 7 | nd |
| Small shield | BE 46131 | Surface metal | nd | 0.5 | nd | 90 | 0.4 | nd | 9 | nd |
| Patina | nd | 1 | nd | 86 | 0.5 | nd | 12 | nd |
| Rim vessel | BE 46147 | Surface metal | 0.3 | 0.1 | 93 | 0.3 | 0.1 | nd | 6 | nd |
| Patina | nd | 2 | nd | 76 | nd | nd | 20 | nd |
| Bottom vessel | BE 46148 | Surface metal | nd | 0.5 | 0.1 | 93 | 0.3 | 0.1 | nd | 6 | nd |
| Patina | nd | 3 | 0.1 | 76 | nd | nd | 20 | nd |

| Table 2. Quantitative results of the chemical analyses performed with EPMA-WDS on metal core. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| EPMA-WDS | ID | Fe | Co | Ni | Cu | Zn | As | Pb | Sn | Sb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Phialae | BE 46152 | 0.2 | 0.1 | 0.1 | 94 | nd | bdl | nd | 6 | bdl |
| BE 46153 | bdl | bdl | bdl | 92 | nd | bdl | bdl | 6 | bdl |
| BE 46149 | 0.1 | nd | bdl | 93 | 0.1 | bdl | 5 | bdl |
| BE 46160 | 0.6 | bdl | bdl | 93 | 0.1 | nd | 6 | bdl |
| BE 46159 | bdl | nd | bdl | 93 | nd | bdl | bdl | 7 | bdl |
| BE 46146 | 0.3 | 0.2 | bdl | 94 | nd | bdl | nd | 5 | bdl |
| BE 46161 | 0.2 | bdl | bdl | 95 | nd | bdl | nd | 5 | bdl |
| Sheet | BE 46145 | 0.2 | bdl | bdl | 85 | nd | bdl | nd | 6 | bdl |
| Ring | BE 45991 | 0.1 | 0.1 | 0.1 | 93 | nd | bdl | nd | 6 | bdl |

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of 0.1–0.3 wt-%. These small amounts of lead could be attributed either to the smelting process since copper ores sometimes contain lead or to its accidental addition through the melting of copper with scrap metal which contained lead (Charalambous 2015). Lead sources in Greece are widespread in Macedonia, Laurion, Cyclades (Vaxevanopoulos 2017) and also in the Pelion area, 30 km southeast from the study area (Teller 1880; Tataris 1960).

Arsenic is in very low concentration in the artefacts and only in a few samples it is present in an amount of 1 wt-%. The low concentration indicates a non-intentional addition in the alloys through the smelting of As-bearing copper ores (Giumlia-Mair 2005) or even through the flux.

Iron is present in all artefacts as a residue from the raw material. The concentrations vary from 0.1 to 2.0 wt-%.

Three samples, coming from a pin, a shaft and a sheet, have a zinc amount of 1–2 wt-%. Could this have happened because the artefacts had the golden colour of the copper–zinc–tin alloy (brass)? (Craddock and Eckstein 2003).

Finally, in a very few cases, there is a presence of Ni. The presence of antimony in two samples can also be attributed to the smelting process (Hauptmann 2007). Nickel concentrations reached 0.3 wt-% and antimony up to 0.4 wt-%.

4. Copper ore sources

The region of Magnesia is characterized by the presence of three distinct sources of copper. Othrys Mount hosts a number of ophiolitic rocks with copper mineralization. It constitutes the most significant copper deposit area. Copper minerals are located in scattered doleritic veins (Koutsovitis 2009). Sulphides such as pyrite have been deposited from hydrothermal fluids (Sovatzoglou-Skounaki 1983). Native copper has been found in a number of altered hydrothermal veins. In southern Othrys, copper metallurgical activity has been testified with a number of slag heaps and debris (Tizzoni et al. 2008).

In Chalchodonion Mount, a copper deposit in the ophiolitic series is located 4 km southwest of ancient Pherae. The ancient term “chalkodonion” of the mountain means the one that gives copper (chalko-). The main copper minerals found on the surface of the mountain are malachite and azurite. No detailed mapping of ore deposition has been done yet.

In Pelion Mount, several deposits have also been found in the northwestern part of the mountain near Zagora village containing mainly galena, sphalerite, pyrite and chalcopyrite (Teller 1880; Tataris 1960; Patousias 1998). Ancient and contemporary mines have been recorded in which minerals such as malachite and azurite are found.

5. Conclusions

Pherae has been proved to be an active production centre with workshops of different natures (Doulgeri-Intzesiloglou 1992, 1994; Asderaki-Tzoumerkioti and Rehren 2006; Orfanou et al. 2014; Orfanou 2015, 2016). Evidence of Hellenistic pottery and lead workshops, as well as an Archaic bronze workshop has been recovered. The aforementioned workshops emphasize a certain tradition in the production of commodities at ancient Pherae.

As has already been presented by Orfanou (2015 and unpublished Ph.D.), the characteristics of careful control of copper-based production in Pherae were exercised. Practical recipes were preferred by the metal-smiths and there was a correlation between object typology and chemical compositions.

The chemical analysis of the assemblage of copper alloy artefacts from the Sanctuary of Hercules in Sesklo in the Pherae area brought to light useful results.

The objects consist of copper alloy and more specifically of low and medium bronze. Metallography revealed that very few objects were produced by casting and they were recognized by their typical microstructure formed of dendrites while the majority of them provided evidence of hot and cold working.

Nickel and antimony are present as trace elements in a very few samples. Arsenic is present in a low percentage of 0.1–0.5 wt-% and this could be attributed either as an impurity of the copper ore or that the metal used was recycled. Iron varies from 0.1 to 3.0 wt-%; however, we cannot safely conclude if this is due to the metal-bearing or due to contamination of corrosion products.

Knowing the potential contribution of pXRF in the analysis of archaeological objects in which in most cases invasive sampling is not the option, and keeping in mind limitations of pXRF one should use the obtained data with caution (Asderaki-Tzoumerkioti and Doulgeri-Intzesiloglou 2010; Asderaki-Tzoumerkioti et al. 2010; Charalambous, Kassianidou, and Rehren 2006; Orfanou et al. 2014; Orfanou 2015, 2016).

A preliminary field study and bibliographical research showed that local copper deposits in the Magnesia area can provide enough material for bronze production.

Further research must be done with an intensive mapping of ore resources in the area, detailed study of ore paragenesis, trace elements and Pb isotopic analysis, which will assure the provenance of the raw material.

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Eleni Asderaki-Tzoumerkioti is an archaeological conservator, based in Greece. She has an MA in Principles in Conservation from UCL, Institute of Archaeology. She worked for 33 years for the Hellenic Ministry of Culture as a senior conservator becoming in 2008 Head of the Department of Conservation of Antiquities in the now-called Ephorate of Antiquities in Magnesia and held this position until her retirement in September 2011. For more than 12 years, she has been collaborating with the IoA UCL and lately with UCL Qatar in various research projects. Understanding the manufacture of ancient metals, pigments and glass is her main research interest.

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