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A contribution to the study of copper production in the Iron Age polity of Paphos in Cyprus

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ABSTRACT

This paper presents the results of the chemical, microscopic and isotopic analysis of an archaeometallurgical assemblage consisting of slag and metal samples from two Cypro-Classical monuments recently located within the urban landscape of the polity of Paphos, and from two slag heaps in the metalliferous region of the Paphos hinterland. The project identified technological changes and innovations, such as the introduction of new types of fluxes and the optimization of the smelting technology. Furthermore, the analysis of slag samples from one of the two monuments, identified as workshop complex on the plateau of the Paphian citadel, revealed the presence of an iron smithy. This pioneering interdisciplinary study paves the way for the development of a comparative archaeo-metallurgical project that will define the fingerprint of the Paphos copper deposits. The study was carried out in the context of the University of Cyprus-Leventis Foundation Project, “From the metalliferous sources to the citadel complex of ancient Paphos: Archaeo-environmental analysis of the mining and the built environment” (acronym MEANING 2017–2019).

1. Introduction

1.1. *‘The tip of the iceberg’*

The contribution of copper resources towards the long-term economic success of the polity that was administered from the primary centre of Paphos on the SW coast, has only recently begun to receive serious consideration (Iacovou 2012). Paphos’ early history (Georgiou 2019), from its foundation in Late Cypriot I (LCI) (1650–1450 BCE) to the termination of its autonomy by Ptolemy I Soter in 310 BCE, was overshadowed by the literary fame of the cult centre of Aphrodite (Iacovou 2019). Since the 1880 s, ‘the sanctuary with the longest unbroken cult tradition in Cyprus’ (Maier 2000) was the focus of archaeological interest, and the study of the sanctuary’s founding polity was confined within the narrow limits of Palaepaphos (Maier 2004, for the history of research). Palaepaphos, however, as the site was known from the 3rd c. BC, to distinguish it from Nea Paphos, was only a sanctuary town; it was no longer the capital centre of the kings of Paphos, who had been in command of an extensive hinterland), rich in water resources from three main rivers and their subsidiaries, timber from the Paphos forest and, almost certainly, copper (Iacovou 2014).

The procurement of copper by the polity of Paphos, and the contribution of local metal resources in the political and economic development of the Paphian region, from the foundation of Paphos in the LBA to the abolition of its autonomous city-state at the end of the 4th c. BC, is one of the research questions that the *Palaepaphos Urban Landscape Project* (hereon PULP) has been at pain to address. Since its initiation in 2006, PULP has carried out targeted excavations that inform the urban structure of the polity in the *longue durée*, and surveys in the cupriferous zone of the Paphos hydrological region/catchment, which is the maximum spatial extent of the project’s landscape analysis. Cyprus’s conventional model of survey units is based on the main and the subsidiary hydrological regions (Peltenburg 2013). According to a study published in 2007 by the Ministry of Agriculture Natural Resources and Environment, Cyprus is divided into nine major hydrological regions (Water Framework Directive 2000/60 EC). The Paphos hydrological

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region (circa 1.173 sq.km.) consists of a group of adjacent hydrological basins, quite large and geomorphologically distinct due to the geological conditions and tectonic processes that have given shape to this unique landscape.

PULP is the first project to approach the establishment of the LC I coastal gateway, and later central place, of Paphos from the polity's economic region. The macro-scale analysis of the 3rd and 2nd millennium BC site distribution in the Paphos catchment suggests a spatial relationship with copper sources (Agapiou et al. 2013). Apparently, at the time of its foundation, Paphos was the terminal link in a chain of industrial establishments and support settlements that originated in the cupriferous pillow lavas on the southern foothills of the Troodos.

Today, as a result of PULP's field work and the application of geospatial analyses, the sanctuary's long biography can be closely associated with the transformations of the urban structure, and the site of Paphos is viewed as a central node between coastal and hinterland landscapes. The presence of ancient copper mines and slag heaps suggests that mining and smelting did take place in the Paphos catchment (Fig. 1). But was copper from these production centres used in the manufacture of objects of high craftsmanship that first appear in the tombs of Paphos at the time of the establishment of the LC precursor of the sanctuary of Aphrodite? Although rarely mentioned, copper slag from metallurgical activity was discovered in association with LC pottery in the lowest layer of the sanctuary. Megaw (1951), to whom we owe this information, does not fail to add that, it 'offers an explanation of the wealth of the ancient city.' Not long before the crisis years of the 12th c. BC (Georgiou 2017), lavish metal artefacts began to be deposited in the LC tombs of Paphos (Catling 1968; Karageorghis 1990). Unlike other, more prominent, LC urban centres that were abandoned and never reoccupied after the 12th c. BC, Paphos developed into a veritable city-state during the transition to the Early Iron Age (Iacovou 2018). The newly established Cypro-Geometric cemeteries of Paphos (Iacovou 2019, fig. 24) are not matched in wealth anywhere else on the island. The publications by Karageorghis (1983) and Karageorghis and Raptou (2014; 2016; 2019) of tombs in the *Skales* and *Plakes* burial sites, show that the dead were interred with significant quantities of metal artefacts. Recent chemical analysis of the bronzes from these burials demonstrates continuous access to tin (Charalambous et al. 2014; Charalambous and Kassianidou 2014).

The access that the rulers of Paphos must have had to metal resources is eloquently portrayed by the mythical personality of the proverbially wealthy king of Cyprus Kinyras, inventor of copper mining and beloved priest of Aphrodite (Franklin 2015). Of all the royal dynasties known to have ruled in the Cypriot city-states, Kinyras was claimed as the forefather of the kings of Paphos, the region that has the poorest copper deposits. By virtue of their descent from Kinyras, the *Kinyradai* were priests of Aphrodite, and in the rich epigraphic corpus of Paphos four kings bear the double title '*basileus* of Paphos and priest of the *wanassa*' (Maier 1989). These *Kinyradai* are now chronologically associated with two Cypro-Classical monuments recently discovered one km east of the emblematic sanctuary of Aphrodite: a workshop complex on the plateau of Hadjiabdoulla (Fig. 2) and a huge tumulus on the nearby hillock of Laona, which was constructed (most likely at the end of the 4th or early in the 3rd c. BC) over an earlier, 5th-century BCE rampart (Iacovou 2021).

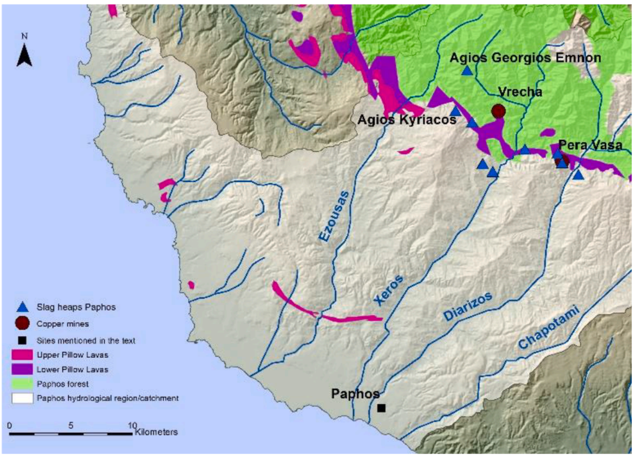

Fig. 1. Map of the Paphos catchment area showing copper mines and slag heaps (Map prepared by V. Kassianidou with digital geological data provided by the Cyprus Geological Survey).

The well-preserved status of the new monuments offered an unprecedented opportunity to explore the resource exploitation strategies and the networking pattern of Paphos in the Cypro-Classical period through a specialised archaeo-environmental project. Launched in 2017, MEANING undertook the interdisciplinary study of the ancient copper slag heaps and the woodland resources of Paphos and carried out analyses of the small archaeometallurgical assemblage from the excavations at Hadjiabdoulla and Laona. Ten years ago, we had noted that 'the survey results of the Gales and Maliotis are, as they themselves write, 'the tip of the iceberg' (Stos-Gale et al. 1998), but they are enough to show that the history of copper extraction in the region of Paphos in antiquity has only just begun' (Iacovou 2012).

1.2. Copper ore deposits, mines, and ancient slag heaps in the Paphos hinterland

In Cyprus, massive sulphide copper ore deposits are located within the geological formation known as the Pillow Lavas (Constantinou 1982) (Fig. 3). The pillow lavas within the Paphos Forest were prospected in 1940 and again in 1970; two ore deposits were identified at Vrecha-Malas and Pera Vasa-Petalas. The orebody at Vrecha-Malas was about 200,000 tonnes and had an average copper content of 0.55 % (Bear 1963; Gass et al. 1994). Because the ore deposit was rather small it was not until 1986 that modern exploitation by open cast started in the area of Vrecha. The mine was active for two years and a total of 11,172 tons of pyrites were extracted (Maliotis 2021). The size of the orebody at Pera Vasa-Petalas was estimated to be about 100,000 tonnes and to have an average copper content of about 0.75 %. The average copper content of these two ore deposits is much lower than that of deposits in other mining regions. In the Mavrovouni ore deposit in the Solea mining region, for example, the average copper content is 4,5% (Adamides 2010; see relevant data in https://www.moa.gov.cy/moa/Mines/MinesSrv.nsf/all/8586AC4EB8686A2EC22574EA0031B407/$file/Copper%20and%20gold%20mines.pdf?openelement)https://www.moa.gov.cy/moa/Mines/MinesSrv.nsf/all/8586AC4EB8686A2EC22574EA0031B407/$file/Copper%20and%20gold%20mines.pdf?openelement.

Ancient slag heaps associated with the copper ore deposits throughout the island are listed in the records of the Geological Survey Department and the Hellenic Mining Company. Several slag heaps of the Paphos catchment were visited and sampled by the Canadian Palaepaphos Survey Project, but none of them were absolutely dated with radiocarbon (Fox et al. 1987). In some of these slag heaps samples were collected that were found to contain manganese, the presence of which offers a relative date for their formation: systematic recording of slag heaps undertaken over the last twenty-five years within the framework of a series of projects enabled the diachronic study of smelting technology on the island and showed that in Cyprus manganese was used as a flux only in the Roman period and in Late Antiquity (Kassianidou 2003a, 2013a, 2021; Socratous et al., 2015; Shaar et al. 2015). A single radiocarbon date for the Pera Vasa heap (H8989-8882), which was published by Zwicker (1986), places the smelting activities there in the 4th c. AD (315 ± 50AD). In 1998, Stos Gale, Maliotis and Gale published a list of known slag heaps around the Troodos Mountains and provided their geographical coordinates and a description of their size in a relative scale ranging from patchy to large. The list includes several small slag heaps in the Paphos Forest and a large slag heap at Agios Kyriacos near

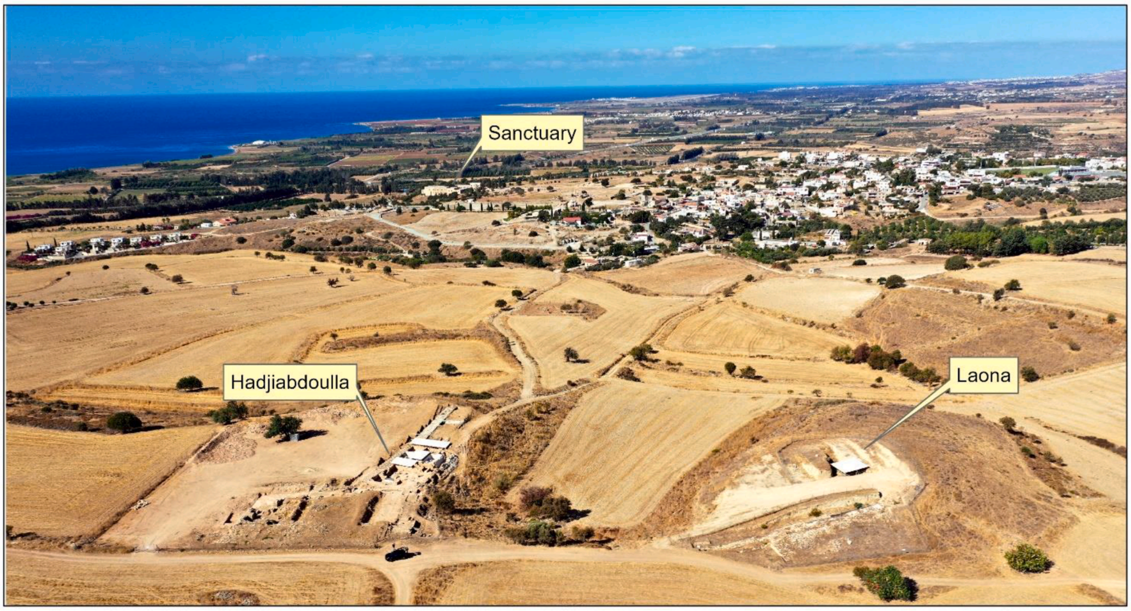

Fig. 2. Oblique aerial view taken over the sites of Palaepaphos Hadjiabdoulla and Laona looking towards the west (Sanctuary) at the height of approximately 100 m above ground level (aerial photo taken by K. Themistocleous, figure compiled by A. Agapiou, the Eratosthenes Research Centre, Cyprus University of Technology ©PULP).

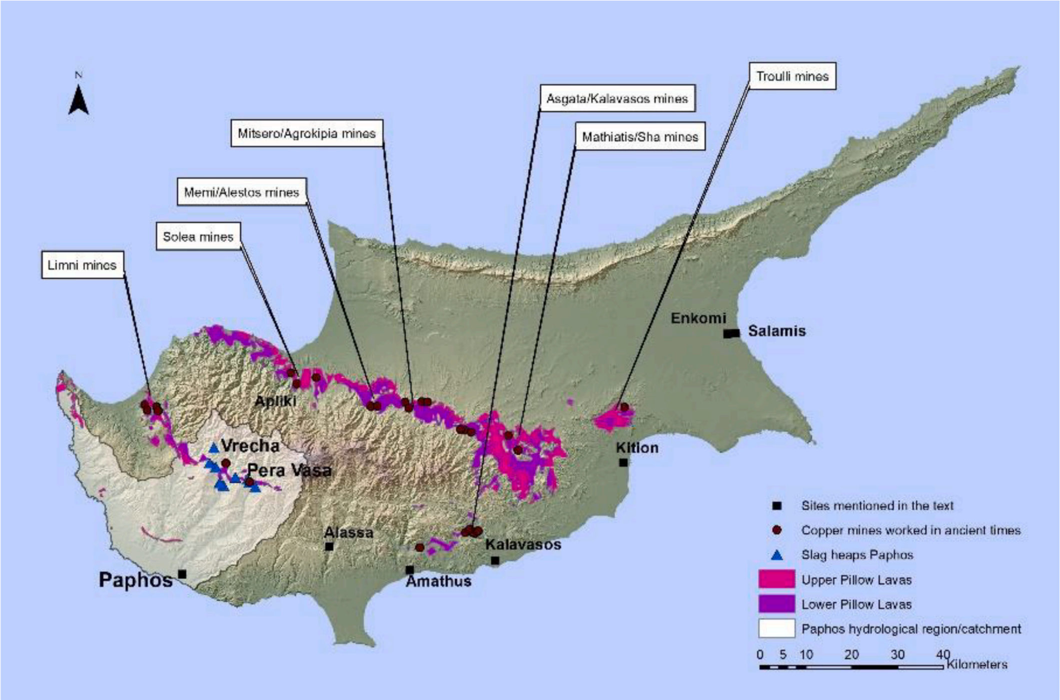

Fig. 3. Map of Cyprus showing the Upper and Lower Pillow Lava formation where the main copper ore deposits are located, the area under study and sites mentioned in the text (Map prepared by V. Kassianidou with digital geological data provided by the Cyprus Geological Survey).

Panagia village (Stos-Gale et al. 1998).

2. Recording the slag heaps in the framework of PULP and MEANING

Within the framework of PULP, eleven slag heaps or slag scatters were georeferenced, described, and where possible their volume was estimated (Iacovou 2014) (Fig. 1). A Leica Viva Global Navigation Satellite System (GNSS) was used to document and georeference the slag heaps and slag scatters, with a few centimetres accuracy, into the national cadastral georeferenced system (Cyprus Geodetic Reference System' 93, Local Traverse Mercator projection). These data were inserted into a Geographical Information System (GIS) and analysed along with the rest of the geodata of PULP. Sampling points were also documented.

The two larger slag heaps were recorded in greater detail in the context of MEANING. The largest of the two is the slag heap of Pera Vasa with a circumference of 100 m. It is hidden under a thick pine forest that was planted after a devastating fire in 1970 (Fig. 4a). The stratigraphy of

Fig. 4. a. The slag heap of Pera Vasa is hidden under a thick pine forest. b. Large slag cake from Pera Vasa slag heap. c. The slag heap of Agios Georgios Emnon. d. Large slag cake from Agios Georgios Emnon slag heap (Photographs V. Kassianidou).

the slag heap was visible in small sections created by the terraces for planting the trees. Large slag cakes 50–60 cm in diameter and more than 10 cm in thickness, which weigh over 40 kilos, are present within the strata and are scattered around the slag heap (Fig. 4b). This suggests that the Pera Vasa slag heap dates to Late Antiquity. Slag cakes of this size have been recorded in the slag heaps of Skouriotissa (Kassianidou, 2013b), Mitsero, Limni (Socratous et al. 2015), Memi (Graham et al. 2006), and Mathiatis (Zwicker 1986); based on associated pottery and radiocarbon dating, they all belong to Late Antiquity (4th – 7th c. A.D.) (Fig. 3). It has therefore been argued that slag cakes of this size can act as markers of copper production dating to Late Antiquity (Kassianidou 2022). Apart from the radiocarbon date published by Zwicker (1986), the chronology of the Pera Vasa slag heap is confirmed by a new radiocarbon date (GU26967), which falls in the 5th – 6th centuries AD (417–560 Cal AD 95 % as calibrated through OxCAL using IntCal 20: Reimer et al. 2020). The largest part of the slag heap, however, is composed of slag that has been crushed into small pieces, a phenomenon that has been observed in all the other Late Antiquity slag heaps. In prehistoric times slag had to be crushed to retrieve the metallic copper prills which were trapped in the matrix. Slag of the Late Antiquity, however, has a very low copper content, often below 1 %, and does not contain large metal prills. This means that it would not have been necessary to crush the slag to retrieve the metal. Why then did they invest so much human energy to do it? It has been argued that this was done to manage the waste and to create stable slag heaps (Kassianidou, 2013b). The strata of the Pera Vasa heap were rich in organic material. Bulk samples were collected in plastic bags and were treated with the water sieve to retrieve the organic material for radiocarbon dating and, more importantly, to identify the species of the trees used to produce the charcoal that fuelled the smelting installations.

The other slag heap that was recorded is below the small church of Agios Georgios Emnon. The heap was formed on a rather steep slope; because there are no large sections, it was impossible to observe the whole stratigraphy and history of smelting activities at this site (Fig. 4c). However, a forest road cut at the bottom of the heap allowed for samples to be taken from stratified deposits. This heap also consists of finely crushed slag, but large slag cakes were also visible (Fig. 4d).

3. The archaeometallurgical assemblage from PULP's excavations at Hadjiabdoulla and Laona

Apart from the samples from the two slag heaps, the study focused on a series of slag and metal samples collected from the excavations of built monuments on Hadjiabdoulla and Laona within the archaeological site of Paphos (Fig. 2). The recent discovery of a 65-meter-long, purpose-built workshop complex attached to the north-west corner of an impressive cut-stone 'palatial' edifice (Maier 2004, 74-76), suggests that the plateau of Hadjiabdoulla had served as the administrative citadel of the city-state of Paphos during the Cypro-Classical period (Iacovou and Karnava 2019). Although the palatial monument, which was only partially excavated in the 1950s, remains unpublished and the excavation of the workshops is still in process, both monuments appear to have been built in the 5th c. BC. Of the units investigated to this day in the workshop complex, seven contain parts of installations (e.g., millstones, stone weights, basins, press beds, drains) associated with the extraction of olive oil (Hadjisavvas 2021), while one was a store for a large quantity of purple shells kept for secondary use after the extraction of the dye (Iacovou and Mylona 2019) (Fig. 5). The small assemblage of slag and metal samples were primarily retrieved from Unit 3, the largest in size (69 sq.m.) single area of the complex, where the crushing and pressing of olives may have continued almost to the end of the 2nd c. BC (Iacovou 2021).

In the case of Laona, the samples come from soil sediments transported to the site for the construction of the huge earthen mound that was raised over an earlier rampart (Gkouma et al. 2021) (Fig. 6). The rampart, or fortress, was also built early in the 5th c. BC (Lorenzon and Iacovou 2019). The tumulus, on the other hand, appears to have been constructed towards the end of the 4th c. or early in the 3rd c. BC. Hence,

Fig. 5. True orthophoto mosaic over the Hadjiabdoulla workshop complex generated from photogrammetric structure from motion (SfM) processing of high-resolution low altitude nadir and oblique drone images. Vectors over the mosaic highlight archaeological findings and trenches (drafted by Athos Agapiou).

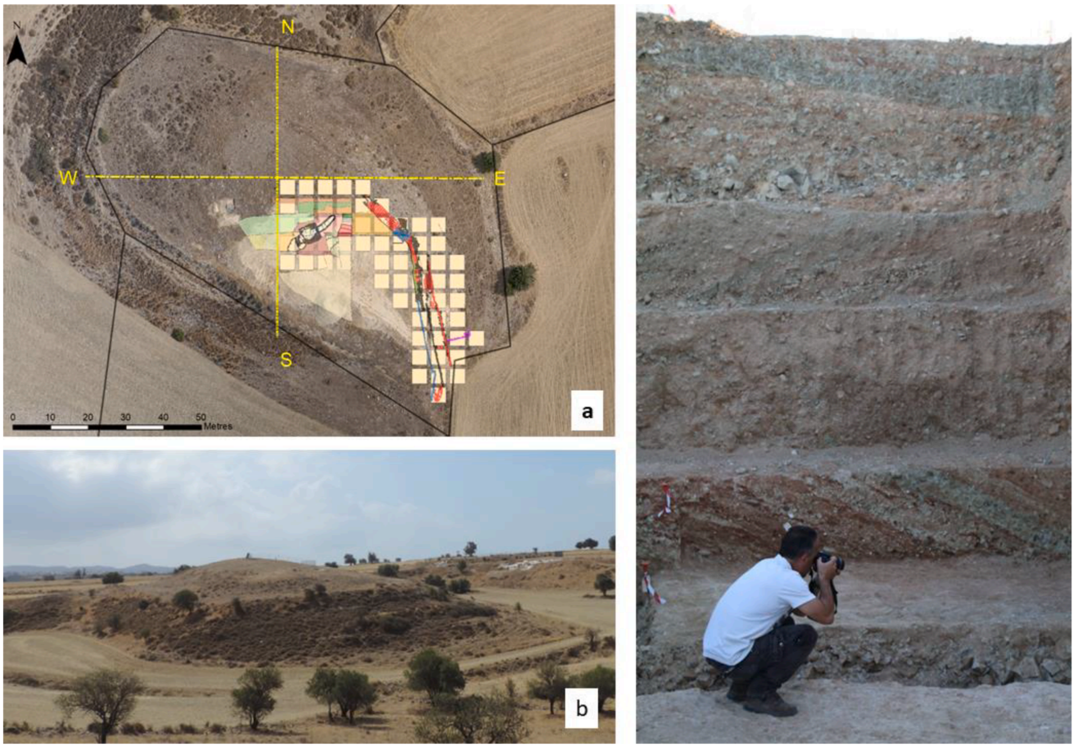

Fig. 6. a. True orthophoto mosaic over laona generated from photogrammetricstructure from motion (SfM) processing of high-resolution low altitude nadir and oblique drone images. Vectors over the mosaic highlight archaeological findings and trenches located in the SE quarter of the mound (drafted by Athos Agapiou). b. Laona from the north before excavation (Photo: M. Iacovou). c. T. Karkanas on Laona, taking photographs of soil sediments exposed against E to W section (Photo: M. Iacovou).

the slag samples from Laona have no secure dating, only a *terminus ante quem* in the 3rd c. BC. Just like the ceramic material collected from the layers of sediments with which the mound was built, the slag may have come from sites dating from as early as the LBA.

3.1. Analytical methodology

A total of 54 slag and 4 metal samples (initially identified as slag samples) were selected for chemical analysis and optical microscopy. The samples are distributed as follows: 21 come from Laona (LA), 8 from Hadjiabdulla (HA), 17 from the Pera Vasa (PV) slag heap and 8 from the Agios Georgios Emnon (AE) slag heap. The chemical analysis was carried out on fresh-cut sections using an Innov-X Delta (now Olympus) handheld portable X-ray Fluorescence Spectrometer (HH-pXRF). The instrument is equipped with a 4 W, 50 kV tantalum anode X-ray tube and a high-performance Silicon Drift Detector (SDD) with a resolution of 150–155 eV (Mo-Kα). The analytical mode of the instrument selected for the slag analysis was “Mining Plus”, while “Alloy Plus” mode (Charalambous et al. 2014, for the analytical parameters”) was applied for the analysis of the 4 metal samples. For “Mining Plus” mode, Beam 1 (40 kV) detects the heavier elements Ti, V, Cr, Mn, Fe, Co,Ni, Cu, Zn, As, Zr, Mo, Pd, Ag, Cd, Sn, Sb, Hf, Ta, W, Pt, Au, Pb, Bi and Beam 2 (10 kV) is used for the determination of Al, Si, P, S, Cl, K and Ca. The diameter of the X-ray beam was 10 mm and the measurement time for each spot analysis was 70 s. The final value reported is the mean value of 3–8 measurements conducted on fresh-cut sections. The data were organised and manipulated in MS Excel and statistically analysed in MatLab. Three certified reference materials have been analysed to assess accuracy and precision of the applied analytical modes. Specifically, two basalt reference materials (BHVO-2 and BCR-2, United States Geological Survey) for "Mining Plus" and a set of copper alloys (BCR-691, European Commission – Joint Research Centre, Belgium) for "Alloy Plus" mode were applied respectively. For all certified reference materials, the measurement precision and accuracy are both very good (coefficient of variation and relative error below 10 % for all oxides and metallic elements).

Polished sections were prepared from all 54 samples for study under a Leica DM2500 P microscope in reflected light. The aim of the microscopic analysis was to examine the microstructure and identify the mineralogical phases of the archaeological slag samples to establish the metallurgical operations taking place at each site and to assess technological aspects, such as the composition of the charge, redox conditions, and the operating temperature of the smelting furnace.

Twenty-four samples of slag and metal from Hadjiabdulla and Laona and the slag heaps in Pera Vasa and Agios Georgios Emnon were selected for Lead Isotope Analysis (hereon LIA) which was carried out at the Centre for Archaeological Sciences of the Department of Earth and Environmental Sciences, KU Leuven. These new analyses make a significant contribution to the isotopic fingerprint of the region, as the OXALID database of LIA of the Oxford Laboratory, includes only 4 ore and 5 slag samples from this area (https://oxalid.arch.ox.ac.uk/The%20Database/TheDatabase.htm). The samples were completely dissolved following a high-temperature acid digestion procedure, from which an aliquot was used for lead isolation and isotope ratio analysis by MC-ICP-MS (Multi-Collector Inductively-Coupled-Plasma Mass Spectrometry) on a Neptune device. Full details of the sample preparation and laboratory procedures are provided in Rademakers et al. (2017). Uncertainties measured are within the currently accepted ranges for LIA, i.e., errors $\sigma < 0.010$ for all ratios in most samples, with slightly higher maximum errors for some. Samples with unusual LIA values and higher errors were re-measured, and LI ratios confirmed.

4. Results

4.1. Chemical analysis

The results of the chemical analysis of the slag samples, performed on fresh-cut sections, are presented in Table 1. The chemical composition of the slag samples from Laona, Pera Vasa and Agios Georgios Emnon is consistent with that of slag deriving from smelting of copper sulphide ores, as they have high Al_2O_3, SiO_2, FeO, CuO and SO_3 concentrations (Bachmann, 1982a; Hauptmann 2020). There are discernible differences in the chemical composition of the slag from the four sites. The differences may reflect changes in the smelting technology across time, or they may be the products of different processes. It is not surprising that copper is one of the elements whose concentration varies significantly. In the samples from Laona copper concentration ranges from 0.4 to 10.5 wt% but in most of the Laona samples it is higher than 0.9 wt% (median 1.0 wt% Cu, average 2.77 wt% Cu). This relatively high concentration of copper is consistent with Cypriot copper smelting slags of the late 2nd and 1st millennium BC, i.e. of the LBA and the Iron Age. On the other hand, 22 out of 25 samples from Pera Vasa and Agios Georgios Emnon have a copper concentration ranging from 0.3 to 0.75 wt% (median 0.5 wt% Cu, average 0.6 wt% Cu) indicating that they are the product of a much more efficient technology typical of Late Antiquity, as shown by the chemical analysis of slags from the area of Skouriotissa and from the area of Mitsero (Kassianidou 2003b; Georgakopoulou and Kassianidou 2013). Surprisingly, the slag samples from Hadjiabdulla were found to have an even lower concentration of copper ($Cu_2O \leq 0.1$ wt%) and a very low concentration of sulphur. This suggests that they are not products of the smelting of copper sulphide ores, typical of Cypriot ore deposits; they are slags related to the processing of iron ores or metallic iron (confirmed by the microscopic analysis below).

Manganese is only present as a trace element in the slag samples from Laona and Hadjiabdulla, but it was detected in very high concentrations (in most of the cases MnO greater than 20 wt%) in most slag samples from Pera Vasa and Agios Georgios Emnon. Five samples from Pera Vasa had a very low concentration of manganese but this is not unexpected. In all the recorded Late Roman heaps of Skouriotissa (Georgakopoulou and Kassianidou 2013) and Mitsero (Kassianidou 2003b) there were slag samples rich in manganese and slag samples poor in manganese. Perhaps in some smelting charges there was no need to add manganese as a flux.

Surprisingly vanadium was detected in the slag samples from both slag heaps, as was a relatively high concentration of nickel, especially in the case of the samples from Agios Georgios Emnon. Nickel is commonly found in Cypriot chalcopyrites (Constantinou 1982; Rapp 1982) but vanadium is not frequently mentioned in the analysis of slag samples. It was detected in chemical analysis of materials found on metallurgical ceramics from Ambelikou-Aletri (Zwicker 1982). A handful of slag samples from a Late Antiquity slag heap at the site Skouriotissa Kitromilia, a smaller slag heap near the well-known one (Georgakopoulou and Kassianidou 2013), also had elevated levels of vanadium but they were an exception rather than the rule, unlike in the samples from the slag heaps of Agios Georgios Emnon and Pera Vasa. Could these trace elements act as markers for the copper produced in Paphos?

The results of the chemical analysis were investigated through the MatLab statistical package to identify possible grouping that could suggest similar origin of the smelting charge or similarities and differences in the production technology. Fig. 7 presents the results of the Principal Components statistical analysis (PCA plot). There is a clear separation of the slag samples from Laona, Hadjiabdulla, Pera Vasa and Agios Georgios Emnon. The slag samples from Pera Vasa create two subgroups, one of which is almost identical to that of Agios Georgios Emnon showing that, although spatially removed, the two smelting workshops were using similar ores and fluxes for the furnace charge and similar smelting technology. A separate group is formed by the 5 samples from Pera Vasa with a low manganese concentration. The group of slag samples from the strata of the Laona tumulus demonstrates a larger variance indicating the diverse provenance of the samples, which may have come from different sites dating to different periods. The slag samples from Hadjiabdulla have a completely different profile reflecting the fact that they are the products of a different process related to iron metallurgy rather than copper metallurgy (see below).

Four samples, 2 from Laona (LA2013.MAA17 and LA2013.MAA19) and 2 from Hadjiabdulla (HA33_3_3 and HA33-HA45_7_1) turned out to be lumps of metal rather than slag. The results of the chemical analysis of the metal samples are presented in Table 2. The two metal samples from Laona are made of copper or copper-based alloy. The first is a lump of pure copper with a high concentration of iron (5,6 wt%) and sulphur (1,8 wt%). The second is a bronze with a tin concentration of 3 wt% and 0.9w% sulphur. In both cases, the presence of sulphur indicates that they are the product of smelting of copper sulphide ores. Sample LA2013.MAA17 can be interpreted as black copper, namely metal that would still need to be refined before being used, while sample LA2013.MAA19 is already alloyed with tin and thus may be casting debris. It is generally accepted that the optimal tin concentration, which balances hardness against brittleness is 10 % but even a lower concentration of tin results in a harder alloy (Scott 1991; Meeks 1993). Extensive analysis of bronzes from all periods in Cyprus undertaken by the authors indicates that alloys of copper with a tin concentration below 5 % are commonly used on the island (Charalambous et al. 2014; Kassianidou and Charalambous 2019; Charalambous et al. 2021), which does not have any tin deposits

Table 1
The results of the chemical analysis of the slag samples.

Sample Number	Inv. Numb.	Al_2O_3	SiO_2	SO_3	K_2O	CaO	TiO_2	MnO	FeO	Cu_2O	V	Cr	Ni	Zn	As
		wt% ± std	wt% ± std	wt% ± std	wt% ± std	wt% ± std	wt% ± std	wt% ± std	wt% ± std	wt% ± std	ppm ± std	ppm ± std	ppm ± std	ppm ± std	ppm ± std
1	LA2012.MAA6	25.0 ± 1.1	21.0 ± 1.0	3.4 ± 0.2	0.4 ± 0.04	0.8 ± 0.05	0.15 ± 0.01	0.03 ± 0.003	39.0 ± 1.2	1.1 ± 0.1	n.d	n.d	n.d	n.d	50 ± 5
2	LA2012.MAA9	16.8 ± 0.9	19.1 ± 1.0	2.0 ± 0.1	0.35 ± 0.03	2.5 ± 0.1	0.1 ± 0.01	0.02 ± 0.002	29.3 ± 1.0	2.5 ± 0.1	n.d	n.d	n.d	n.d	40 ± 5
3	LA2012.MAA7	24.5 ± 1.1	23.5 ± 1.0	3.7 ± 0.2	0.5 ± 0.05	1.4 ± 0.1	0.1 ± 0.01	0.025 ± 0.002	44.2 ± 1.3	0.8 ± 0.05	n.d	n.d	n.d	n.d	60 ± 5
4	LA2013.MAA6	24.7 ± 1.1	28.7 ± 1.3	1.9 ± 0.1	0.7 ± 0.05	1.1 ± 0.1	0.1 ± 0.01	0.05 ± 0.005	35.8 ± 1.5	6.8 ± 0.5	n.d	n.d	n.d	n.d	140 ± 10
5	LA2013.MAA4	26.2 ± 1.2	27.2 ± 1.2	1.9 ± 0.1	0.3 ± 0.03	1.4 ± 0.1	0.2 ± 0.02	0.04 ± 0.004	32.9 ± 1.3	5.9 ± 0.4	n.d	n.d	n.d	n.d	130 ± 10
6	LA2013.MAA7	25.0 ± 1.0	26.5 ± 1.1	3.2 ± 0.2	0.5 ± 0.05	n.d	0.15 ± 0.01	0.02 ± 0.002	40.0 ± 1.4	1.0 ± 0.1	n.d	n.d	n.d	n.d	60 ± 5
7	LA2013.MAA8	25.4 ± 1.2	23.5 ± 1.0	1.8 ± 0.1	0.5 ± 0.05	n.d	0.15 ± 0.01	0.03 ± 0.003	43.5 ± 1.4	0.9 ± 0.05	n.d	130 ± 10	n.d	n.d	60 ± 5
8	LA2013.MAA10	20.0 ± 0.8	18.9 ± 0.9	1.8 ± 0.1	0.6 ± 0.05	0.9 ± 0.05	0.1 ± 0.01	0.035 ± 0.003	30.2 ± 1.2	0.4 ± 0.04	n.d	n.d	n.d	n.d	50 ± 5
9	LA2013.MAA11	15.7 ± 0.7	8.8 ± 0.5	8.8 ± 0.5	0.1 ± 0.01	2.0 ± 0.1	n.d	n.d	25.6 ± 1.0	6.7 ± 0.4	n.d	n.d	n.d	n.d	n.d
10	LA2013.MAA12	16.1 ± 0.7	19.9 ± 0.9	0.9 ± 0.05	0.25 ± 0.02	n.d	0.1 ± 0.01	n.d	27.5 ± 1.0	1.0 ± 0.1	n.d	n.d	80 ± 5	n.d	30 ± 5
11	LA2013.MAA13	15.4 ± 0.7	17.1 ± 0.8	2.0 ± 0.1	0.3 ± 0.03	0.4 ± 0.04	0.07 ± 0.005	0.01 ± 0.001	22.8 ± 1.0	1.0 ± 0.1	n.d	n.d	60 ± 5	n.d	30 ± 5
12	LA2013.MAA18	24.0 ± 1.2	22.7 ± 1.1	2.8 ± 0.1	0.5 ± 0.05	2.8 ± 0.2	0.1 ± 0.01	0.03 ± 0.003	45.0 ± 1.5	1.0 ± 0.1	n.d	n.d	n.d	n.d	70 ± 5
13	LA2013.MAA19	20.0 ± 0.8	27.7 ± 1.1	0.5 ± 0.05	1.0 ± 0.1	4.3 ± 0.3	0.2 ± 0.02	0.02 ± 0.002	33.5 ± 1.1	10.5 ± 0.5	n.d	n.d	n.d	n.d	440 ± 30
14	LA2013.MAA5	20.3 ± 0.8	20.4 ± 1.0	3.5 ± 0.2	0.5 ± 0.05	n.d	0.1 ± 0.01	0.02 ± 0.002	33.5 ± 1.2	1.1 ± 0.1	n.d	n.d	120 ± 10	n.d	n.d
15	LA2013B.MAA6	22.9 ± 0.8	19.3 ± 1.0	2.5 ± 0.2	0.3 ± 0.03	0.6 ± 0.05	0.07 ± 0.005	0.015 ± 0.001	32.5 ± 1.1	4.2 ± 0.3	n.d	n.d	n.d	n.d	190 ± 10
16	LA11_1_1	20.6 ± 0.8	19.0 ± 1.0	1.8 ± 0.1	0.35 ± 0.03	1.0 ± 0.1	0.1 ± 0.01	0.02 ± 0.002	35.5 ± 1.2	0.4 ± 0.04	n.d	n.d	n.d	n.d	50 ± 5
17	LA30_2_1	24.0 ± 1.2	23.2 ± 1.1	5.0 ± 0.3	0.4 ± 0.04	n.d	0.15 ± 0.01	0.02 ± 0.002	44.2 ± 1.5	2.2 ± 0.1	n.d	n.d	n.d	n.d	60 ± 5
18	LA30_2_2	25.7 ± 1.0	26.7 ± 1.2	3.2 ± 0.2	0.4 ± 0.04	0.7 ± 0.05	0.1 ± 0.01	0.025 ± 0.002	40.0 ± 1.3	1.0 ± 0.1	n.d	0.01	160 ± 10	n.d	70 ± 5
19	LA2016.MAA20	25.3 ± 1.1	27.0 ± 1.2	3.1 ± 0.2	0.6 ± 0.05	n.d	0.2 ± 0.02	0.04 ± 0.004	38.0 ± 1.3	1.0 ± 0.1	n.d	n.d	160 ± 10	n.d	70 ± 5
20	LA2016.MAA7	23.0 ± 0.8	24.7 ± 1.1	1.8 ± 0.1	0.5 ± 0.05	1.3 ± 0.1	0.2 ± 0.02	0.02 ± 0.002	34.3 ± 1.2	0.4 ± 0.04	n.d	n.d	170 ± 10	n.d	50 ± 5
21	LA2017.MAA002	22.1 ± 0.8	22.3 ± 0.9	7.2 ± 0.5	0.4 ± 0.04	1.4 ± 0.1	0.1 ± 0.01	0.015 ± 0.001	38.8 ± 1.2	6.2 ± 0.3	n.d	150 ± 10	n.d	n.d	n.d
22	HA33_5_2 – (1)	23.2 ± 0.9	15.0 ± 0.8	0.2 ± 0.02	0.1 ± 0.01	5.3 ± 0.4	n.d	0.06 ± 0.005	47.5 ± 1.4	0.06 ± 0.005	n.d	n.d	310 ± 30	n.d	130 ± 10
23	HA33_5_2 – (2)	23.6 ± 0.8	17.0 ± 0.8	0.3 ± 0.03	0.1 ± 0.01	6.7 ± 0.4	0.08 ± 0.005	0.07 ± 0.005	46.5 ± 1.3	0.06 ± 0.005	n.d	n.d	270 ± 20	n.d	130 ± 10
24	HA33_5_4 – (1)	23.1 ± 0.9	18.9 ± 0.9	0.3 ± 0.03	0.1 ± 0.01	5.0 ± 0.4	n.d	0.05 ± 0.005	50.6 ± 1.5	0.1 ± 0.01	n.d	n.d	550 ± 50	n.d	190 ± 10
25	HA33_5_4 – (2)	24.3 ± 1.0	17.2 ± 0.9	0.3 ± 0.03	0.1 ± 0.01	4.5 ± 0.3	n.d	0.03 ± 0.003	52.2 ± 1.4	0.1 ± 0.01	n.d	n.d	490 ± 50	n.d	210 ± 10
26	HA33_5_4 (no23)	25.0 ± 1.0	5.5 ± 0.3	0.3 ± 0.03	0.1 ± 0.01	n.d	n.d	n.d	48.3 ± 1.3	0.06 ± 0.005	n.d	n.d	250 ± 20	n.d	240 ± 20
27	HA33_5_5 – (1)	30.1 ± 1.3	9.1 ± 0.4	0.3 ± 0.03	0.1 ± 0.01	n.d	n.d	0.04 ± 0.003	54.5 ± 1.5	0.07 ± 0.005	n.d	n.d	490 ± 50	n.d	190 ± 10
28	HA33_5_5 – (2)	22.1 ± 0.8	19.0 ± 0.8	n.d	0.25 ± 0.02	1.3 ± 0.1	n.d	0.15 ± 0.01	36.4 ± 1.2	0.06 ± 0.005	110 ± 10	n.d	0.03	n.d	70 ± 5
29	HA34_5_1	24.2 ± 1.0	18.3 ± 0.8	0.4 ± 0.03	0.1 ± 0.01	6.2 ± 0.3	0.07 ± 0.005	0.4 ± 0.04	47.7 ± 1.2	0.07 ± 0.005	n.d	n.d	490 ± 50	n.d	160 ± 10

(*continued on next page*)

Table 1 (*continued*)

Sample Number	**Inv. Numb.**	Al_2O_3 wt% ± std	SiO_2 wt% ± std	SO_3 wt% ± std	K_2O wt% ± std	CaO wt% ± std	TiO_2 wt% ± std	MnO wt% ± std	FeO wt% ± std	Cu_2O wt% ± std	V ppm ± std	Cr ppm ± std	Ni ppm ± std	Zn ppm ± std	As ppm ± std
30	PV Point 1 31a	6.2 ± 0.3	25.8 ± 0.9	2.2 ± 0.1	0.4 ± 0.04	2.8 ± 0.1	0.3 ± 0.03	20.0 ± 0.8	8.2 ± 0.4	3.5 ± 0.2	1300 ± 50	n.d	620 ± 50	n.d	n.d
31	PV Point 1–31b	10.0 ± 0.4	32.4 ± 1.2	1.9 ± 0.1	0.6 ± 0.05	1.1 ± 0.1	0.3 ± 0.03	20.0 ± 0.8	11.2 ± 0.5	1.9 ± 0.1	1000 ± 50	n.d	820 ± 50	n.d	n.d
32	PV Point 1–31c	13.3 ± 0.4	29.8 ± 1.3	2.8 ± 0.2	0.7 ± 0.05	4.7 ± 0.3	0.3 ± 0.03	24.6 ± 0.9	8.0 ± 0.4	0.5 ± 0.04	1200 ± 50	n.d	700 ± 50	n.d	n.d
33	PV Point 2–32a	8.7 ± 0.3	30.0 ± 1.2	3.0 ± 0.2	0.8 ± 0.05	6.7 ± 0.4	0.4 ± 0.04	23.3 ± 0.9	6.8 ± 0.3	0.5 ± 0.05	1300 ± 50	n.d	510 ± 50	n.d	n.d
34	PV Point 2–32b	25.8 ± 1.0	27.2 ± 1.1	1.8 ± 0.1	0.2 ± 0.02	3.7 ± 0.2	0.3 ± 0.03	0.6 ± 0.05	34.7 ± 1.2	0.6 ± 0.05	550 ± 30	n.d	n.d	800 ± 50	70 ± 5
35	PV Point 2–32c	6.5 ± 0.3	27.5 ± 1.1	1.9 ± 0.1	0.6 ± 0.05	5.0 ± 0.4	0.3 ± 0.03	21.3 ± 0.8	5.1 ± 0.3	0.35 ± 0.03	790 ± 50	n.d	450 ± 50	n.d	n.d
36	PV Point 3–33a	8.3 ± 0.3	29.2 ± 1.2	1.8 ± 0.1	0.8 ± 0.05	4.5 ± 0.3	0.3 ± 0.03	24.6 ± 0.9	9.0 ± 0.5	0.4 ± 0.04	760 ± 50	n.d	780 ± 50	n.d	n.d
37	PV Point 3–33b	7.8 ± 0.3	30.3 ± 1.3	1.8 ± 0.1	0.8 ± 0.05	4.5 ± 0.3	0.3 ± 0.03	24.6 ± 0.9	7.3 ± 0.4	0.3 ± 0.03	780 ± 50	n.d	620 ± 50	n.d	30 ± 5
38	PV Point 3–33c	9.9 ± 0.4	31.7 ± 1.2	2.0 ± 0.1	0.9 ± 0.05	4.6 ± 0.3	0.3 ± 0.03	27.9 ± 1.0	8.2 ± 0.4	0.35 ± 0.03	900 ± 50	n.d	730 ± 50	n.d	40 ± 5
39	PV Point 4–34a	24.0 ± 1.1	30.8 ± 1.3	1.7 ± 0.1	0.7 ± 0.05	4.4 ± 0.3	0.15 ± 0.01	0.2 ± 0.02	37.7 ± 1.2	0.4 ± 0.04	270 ± 20	n.d	n.d	1400 ± 100	70 ± 5
40	PV Point 4 – 34b	23.8 ± 1.0	30.6 ± 1.2	1.8 ± 0.1	0.6 ± 0.05	3.8 ± 0.3	0.15 ± 0.01	0.6 ± 0.05	38.9 ± 1.2	0.6 ± 0.05	410 ± 30	n.d	200 ± 20	1800 ± 100	70 ± 5
41	PV Point 4–34c	25.5 ± 1.1	32.7 ± 1.4	1.6 ± 0.1	0.3 ± 0.03	5.3 ± 0.4	0.3 ± 0.03	0.2 ± 0.02	33.2 ± 1.1	0.45 ± 0.04	280 ± 20	n.d	n.d	700 ± 50	50 ± 5
42	PV – 2022.1	9.6 ± 0.4	26.7 ± 1.1	2.0 ± 0.1	0.4 ± 0.04	5.5 ± 0.4	0.4 ± 0.04	28.4 ± 0.9	9.2 ± 0.5	0.75 ± 0.05	1700 ± 100	n.d	650 ± 50	n.d	n.d
43	PV – 2022.2	8.2 ± 0.3	29.6 ± 1.2	2.0 ± 0.1	0.4 ± 0.04	5.0 ± 0.4	0.3 ± 0.03	28.9 ± 1.0	11.8 ± 0.5	0.75 ± 0.05	1100 ± 50	n.d	910 ± 50	n.d	40 ± 5
44	PV – 2022.3	26.3 ± 1.0	30.5 ± 1.2	2.0 ± 0.1	0.5 ± 0.04	5.1 ± 0.4	0.15 ± 0.01	0.5 ± 0.05	33.2 ± 1.1	0.6 ± 0.05	270 ± 20	n.d	140 ± 10	1000 ± 50	90 ± 5
45	PV – 2022.4	15.9 ± 0.5	34.4 ± 1.1	2.4 ± 0.1	0.4 ± 0.04	6.5 ± 0.4	0.4 ± 0.04	28.0 ± 0.9	12.5 ± 0.5	0.5 ± 0.05	1000 ± 50	n.d	930 ± 50	n.d	40 ± 5
46	PV – 2022.5	15.3 ± 0.5	33.5 ± 1.1	2.5 ± 0.1	0.3 ± 0.03	6.7 ± 0.4	0.45 ± 0.04	27.8 ± 0.9	10.4 ± 0.5	0.6 ± 0.05	2100 ± 100	n.d	720 ± 50	n.d	30 ± 5
47	AE – 35a (Point 1)	10.9 ± 0.4	30.4 ± 1.2	1.4 ± 0.1	0.7 ± 0.05	4.6 ± 0.3	0.3 ± 0.03	14.8 ± 0.5	12.6 ± 0.5	0.4 ± 0.04	1000 ± 50	n.d	760 ± 50	n.d	30 ± 5
48	AE – 35b (Point 1)	9.5 ± 0.4	29.7 ± 1.2	1.6 ± 0.1	1.0 ± 0.1	4.3 ± 0.3	0.4 ± 0.03	22.0 ± 0.8	9.1 ± 0.4	0.4 ± 0.04	1100 ± 50	n.d	750 ± 50	n.d	40 ± 5
49	AE Point 1 – 35c	7.3 ± 0.3	28.3 ± 1.1	1.3 ± 0.1	0.6 ± 0.05	3.6 ± 0.3	0.3 ± 0.03	18.2 ± 0.7	5.7 ± 0.3	0.3 ± 0.03	1100 ± 50	n.d	450 ± 50	n.d	n.d
50	AE Point 1–35d	10.4 ± 0.4	30.1 ± 1.2	1.6 ± 0.1	1.0 ± 0.1	4.3 ± 0.3	0.4 ± 0.03	22.7 ± 0.8	9.5 ± 0.4	0.4 ± 0.04	1300 ± 50	n.d	850 ± 50	n.d	40 ± 5
51	AE Point 2 – 36a	9.4 ± 0.4	32.9 ± 1.3	2.9 ± 0.2	0.7 ± 0.05	3.3 ± 0.2	0.4 ± 0.03	25.5 ± 1.0	17.7 ± 0.6	0.7 ± 0.05	1400 ± 50	n.d	1500 ± 100	n.d	50 ± 5
52	AE Point 2–36b	19.9 ± 0.7	33.7 ± 1.5	1.5 ± 0.1	0.9 ± 0.05	4.1 ± 0.3	0.3 ± 0.03	14.4 ± 0.7	24.4 ± 0.8	0.5 ± 0.04	1000 ± 50	n.d	1400 ± 100	n.d	50 ± 5
53	AE Point 2 – 36c	14.0 ± 0.5	35.5 ± 1.3	1.5 ± 0.1	0.7 ± 0.05	3.2 ± 0.2	0.3 ± 0.03	14.0 ± 0.7	15.8 ± 0.6	0.3 ± 0.03	1000 ± 50	n.d	1000 ± 100	120 ± 10	n.d
54	AE Point 3–37	n.d	22.5 ± 0.9	0.4 ± 0.04	0.4 ± 0.04	n.d	0.2 ± 0.02	28.5 ± 1.1	7.1 ± 0.4	2.1 ± 0.1	1700 ± 100	n.d	1000 ± 100	n.d	n.d

(Constantinou 1982).

In the case of the 2 metal samples from Hadjiabdulla, the first object is made of iron (the concentration of iron is relatively low because the sample is highly corroded), while the second is a copper alloy with high concentration of tin. When the amorphous lump was sectioned, two different phases were discernible each of which had a different colour: one was red and the other was golden yellow. These were analyzed separately. The analysis showed that the red phase had a tin concentration of around 11 % (10.9 ± 0.5 wt% Sn), while the golden phase had a higher tin concentration of around 15 wt% (15.1 ± 0.5 wt% Sn). Such high tin concentrations have been previously detected in the assemblage of metal artefacts from Palaepaphos (Charalambous et al. 2014; Charalambous and Kassianidou 2014), as well as Enkomi (Charalambous et al. 2021) and Salamis (Kassianidou and Charalambous 2019). Bronzes with a high tin concentration were mostly used to produce vessels, particularly bowls of the hemispherical type, which are characteristic products of Cypriot workshops since the LBA. This find may represent different masses of metal that were melted together (perhaps they were being recycled) but did have the time to completely fuse together.

4.2. *Microscopic analysis*

The microscopic analysis of the slag and metal samples made a significant contribution to our understanding of the metallurgical processes represented by their waste and final products. All samples from the two heaps demonstrate a characteristic flow texture indicating that the slag was tapped. The main mineral phases are silicates and sulphide inclusions embedded in a glassy matrix (Fig. 8). The silicate mineral phases, which dominate the microstructure of the slag, consist of large laths of light to medium grey crystals measuring up to 1.5 mm. Variations in the size and thickness are attributed to differential cooling rates. The silicates have precipitated from an iron-manganese silicate melt and are identified as olivines with intermediate compositions between tephroite and fayalite. Interestingly, a small number of slag samples

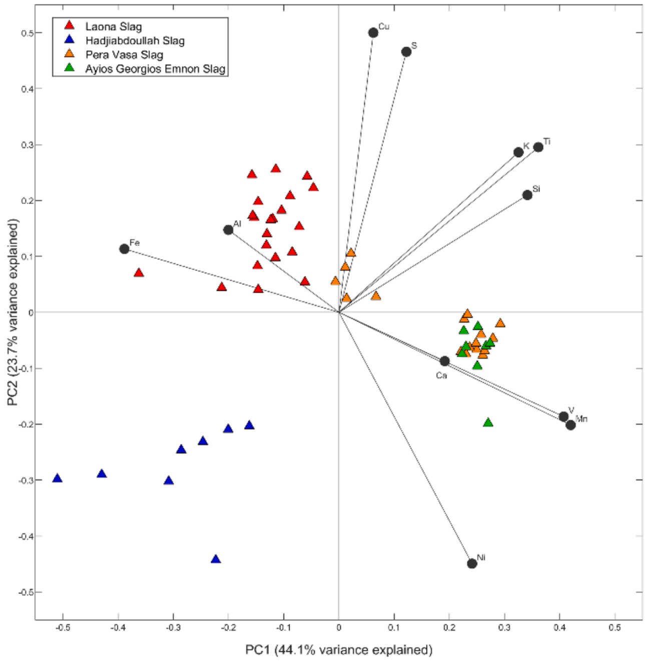

Fig. 7. PCA plot showing the different chemical compositions of slag samples from the four sites Laona, Hadjiabdulla, Pera Vasa and Agios Georgios Emnon (Drawn by Demetrios Ioannides).

Table 2
The results of the chemical analysis of the metal samples.

Sample Number	Inventory Number	Chemical composition (wt% ± std)					
		Cu	Sn	Pb	Fe	As	S
55	LA2013. MAA17	92.6 ± 0.5	n.d	n.d	5.6 ± 0.2	n.d	1.8 ± 0.1
56	LA2013. MAA19	95.4 ± 0.5	3.0 ± 0.2	0.18 ± 0.1	0.5 ± 0.03	n.d	0.9 ± 0.05
57	HA33_3_3	0.07 ± 0.005	n.d	n.d	25.4 ± 0.8	0.03 ± 0.003	0.1 ± 0.01
58	HA33-HA45_7_1 – (Gold metal area)	84.5 ± 0.8	15.1 ± 0.5	0.015 ± 0.001	0.3 ± 0.03	n.d	n.d
58	HA33-HA45_7_1 – (Red area)	88.9 ± 0.7	10.9 ± 0.5	n.d	0.2 ± 0.02	n.d	n.d

from Pera Vasa were crystallized from a Mn-poor iron silicate melt. The excess of trivalent (Fe^{3+}) iron in these samples, introduced by the charge, reacts in a confined manner with the silicate melt, which consumes most of the divalent (Fe^{2+}) iron in the form of fayalites, and solidified as free iron oxides (Fig. 9). A thin magnetite skin defines different tapping layers (Fig. 9b).

Iron-copper sulphides appear mostly as small, irregular phases no larger than 100 μm, dispersed within the glassy matrix (Fig. 8a). These have previously been interpreted as crushed unreacted ore (Kassianidou 2003b). There are also rounded prills of max. 500 μm in size and larger amorphous inclusions, which range from 100 to 500 μm (Fig. 8b). Optically these inclusions vary from the most frequent phase of yellowish iron-rich sulphide, which often demonstrates a bright yellow phase in the centre of the feature to dark yellow, orange, and pink phases which contain higher levels of copper. The latter usually occur in the larger phases, which display a zoning of Fe-rich to Cu-rich compositions from the centre to the outer parts of the inclusions.

The microscopic study is consistent with the chemical analysis; it shows that the slag samples are derived from two technologically distinct metallurgical operations. In the first, iron and silica-rich minerals were probably used as flux. The origin of iron and silica-rich minerals must be sought either in the ochreous sediments (gossans) which cap the ore bodies and contain significant proportions of iron (hydr)oxides and silica (Constantinou 1980, 1982), or in the massive sulphide mineralization which is sometimes associated with amorphous silicon dioxide, but also microcrystalline silica minerals such as opal and jasper (Bear 1963; Adamides 2010; Antivachis 2015).

Some of the slags, however, derive from the primary smelting of copper sulphide ores together with manganese-rich minerals which were added as flux. The use of Mn oxides as fluxing agents is known to offer numerous advantages and was already used in early smelting operations in Faynan (see Hauptmann 2007 and references therein). These manganese rich slags are the waste products of a technologically more advanced smelting operation (Bachmann 1982b), which has been shown to date to Late Antiquity. The manganese rich minerals can be found in the sporadic outcrops of umber, a sediment consisting of iron hydroxides, oxyhydroxides and considerable amounts of manganese, which are stratigraphically separated from the sulphide ore body (Constantinou 1980). However, the existence of self-fluxing sulphide ores containing umber should also be considered since such ores have been recorded within the ore body of Skouriotissa (Kortan 1970).

The microscopic analysis of the samples from Laona revealed that the slags were tapped from the smelting installations in several batches as indicated by the spinifex texture spotted across the assemblage and the presence of multiple tapping layers usually defined by a thin magnetite skin. The major components of all the samples of tap slag are iron silicates, iron oxides and sulphides embedded in a glassy matrix, occurring in variable amounts and compositions (Fig. 10). The silicate mineral phases, which dominate the microstructure of the slag, primarily consist of large laths of light to medium grey crystals measuring up to 1.5 mm. Based on the results of the chemical analysis, the silicates have precipitated from an iron-rich melt with very low calcium content. Thus, they can be described as orthosilicates with a chemical formula approximating the Fe-rich end member of the olivine group of fayalites. Fe-oxides occur consistently in rather invariably increased quantities in the entirety of the Laona slag assemblage. The dominant oxide occurrences are dendrites and/or fine elongated globules of wüstite, while larger grains of magnetite and hercynite-magnetite intergrowths are less commonly encountered. The dominance of wüstites and the presence of hercynite-magnetite spinels suggest that the slag was formed in a smelting process applying rather strong reducing, redox conditions.

The main occurrence of iron-copper-sulphides is in the form of small, irregular, yellowish iron-rich phases of max. 100 μm in size; they have been identified as unreacted crushed ore (Kassianidou 2003b). Larger inclusions and prills with a higher copper content are infrequently present. A single microstructural outlier (LA-14), which is distinguished from the rest of the assemblage, is characterized by increased abundancies of magnetite grains, copper sulphides and metallic copper prills. This sample points to a different metallurgical operation, which was carried out under milder redox conditions, and involves the conversion of matte to raw metal and/or the refining of the latter to pure metal.

The Laona slag samples were produced from the primary smelting of copper sulphide ores, in which the product was a copper rich matte. The addition of fluxes cannot be confirmed since the Cypriot type ore deposits especially in the higher, secondary zones contain significant amounts of silica and iron (hydr)oxides (both spotted in the slag), which may mean that the ores were self-fluxing. Importantly, secondary operations, such as matte conversion or refining/re-melting operations of raw copper have also been identified in the Laona samples. This suggests that the small pieces of slag from the strata of the tumulus represent the entire sequence of copper production from sulphide ores.

The slag samples from Hadjiabdulla have a completely different microstructure, which leads to the conclusion that they are the waste

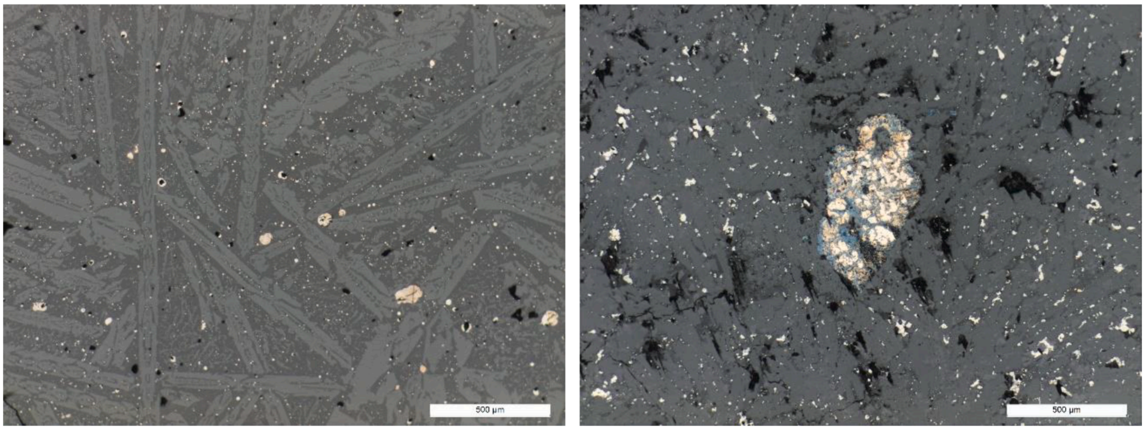

Fig. 8. Photomicrographs (mag. 10x) a: sample AE-35a and b: sample PV-32a showing the general microstructure of the slag. Well-shaped olivine crystals accompanied by small copper-iron sulphides and matte prills.

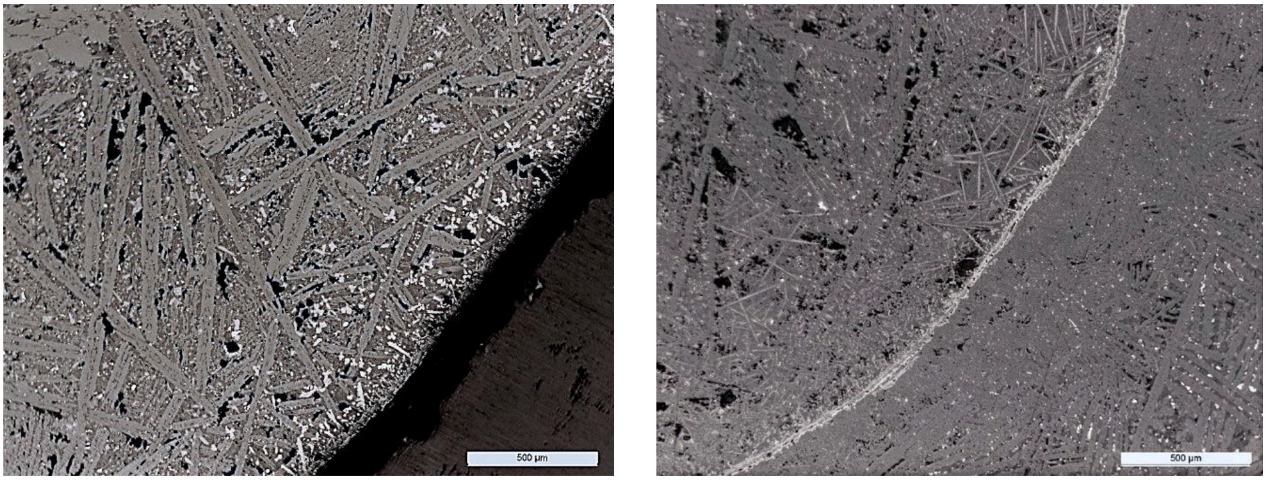

Fig. 9. Photomicrographs (mag. 10x) a: sample PV-34a showing the general microstructure: olivine laths (medium grey) and iron oxide angular and skeletal grains (light grey) embedded in a glassy matrix (black) and b: sample PV-34b (mag. 10x) showing different tapping layers. Note the thin magnetite band that marks out the various tapping layers.

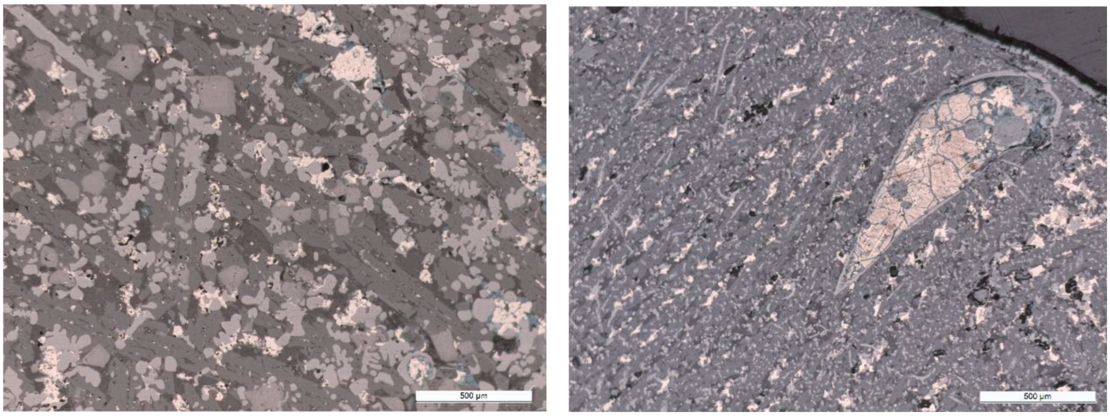

Fig. 10. Photomicrographs (mag. 10x) a: sample LA-11 showing the general microstructure of the slag: medium grey olivine laths, light grey dendrites and elongated globules of wüstite, larger, angular Fe-(Al)-rich particles and iron-copper (yellow) and copper (blue) sulphides. b: LA-01 a large matte prill, needle-shaped silicate laths (medium grey) solidify perpendicularly to the edge of the slag (spinifex texture). The same orientation is observed for the iron oxides (light grey) and the Fe-rich sulphides.

product of a different metallurgical process. The main mineral phases observed in these samples are iron oxides, iron silicates, a glassy matrix and iron. The lighter, yellowish grey colour of the iron oxides distinguishes the crystallisation of wüstite (FeO) from magnetite (Fe_3O_4) which has a more brownish tint (Bachmann 1982a). Wüstite habit varies from dendritic, to hopper shaped to more subhedral, compacted crystals.

Additionally, most of the samples exhibit patches of iron oxide in which the wüstite crystal edges are no longer well defined as they form anhedral crystals known as hammer scale texture (Fig. 11). The texture of the slag is rather homogeneous, indicating a rather constant hearth temperature. These temperatures must have been elevated since dendrites are formed. The dendrites and the few hopper shaped wüstites represent fast cooling, but not quenching: these forms usually occur when the metal is taken out of the hearth and cools immediately to room temperature (Bauvais 2007). The iron oxides in most of the samples are heavily compacted and often form hammer scales. These accumulations of wüstite are partially dissolved fragments of the oxidised crust of iron knocked off during hammering in the hearth (Serneels and Perret 2003; Eekelers et al. 2016). Probably these slags, with the limited slag material (silicates) and the dominant iron oxides, were formed during bar smithing, in which frequent hammering is necessitated to compact the bloom to a bar (Crew 1991). Therefore, it is suggested that secondary iron smithing operations were taking place in the Hadjiabdoulla workshop complex. Iron bloom must have been produced somewhere else in the area (based on the LIA analysis) and was processed further on site.

It has long been known that Cyprus was one of the first areas of the eastern Mediterranean to smelt and use iron (Snodgrass 1982) sometime in the 12th century BCE. In fact, it has been argued that the island played a leading role in the development and dissemination of iron metallurgy (Sherratt 1994; Muhly 2003). The discovery, therefore, of iron smithy slags in the workshop complex of the citadel, which was operating during the 5th and 4th centuries BC, is in no way extraordinary. It is, however, extremely valuable because up to now material remains of ancient iron processing in Cyprus are rare. The only other known iron smithy was found in a roughly contemporary workshop complex of the 4th-3rd centuries BC that was found on the Ayios Georgios hill in the heart of modern-day Nicosia/Lefkosia (Pilides et al. 2007). Despite the absence of epigraphic confirmation, the site of Ayios Georgios is unanimously considered as the location of the little-known Cypriot polity of Ledra (Pilides 2018).

4.3. *Lead isotope analysis*

Twenty-four samples of slag and metal were selected for LIA. The original set up was to compare the isotopic fingerprint of the samples collected in the excavated sites of Hadjiabdulla and Laona with those from the two slag heaps in the catchment area of Paphos and with data in the OXALID database. The results of the analysis are shown in Table 3 and illustrated in Fig. 12 and Fig. 13. The study of the lead isotope fingerprint of Cypriot copper ore deposits published in 1997 did not include any samples from the two ore deposits at Pera Vasa-Petalas and Vrecha-Malas (Gale et al. 1997). However, it included samples from the ore deposits of the area of Limni, which lies to the north west in a different catchment (Gale et al. 1997, Fig. 2) (Fig. 3). Moreover, the online OXALID database includes four ores from Vrecha and Pera Vasa, which have a composition similar to the Limni ores, and thus could be considered part of the Limni Axis.

The LIA of the slags from Agios Georgios Emnon shows a partial overlap with the Limni axis ore samples, as do some of the samples from Pera Vasa (Fig. 12). Other samples from Pera Vasa, however, display a very different isotopic signature, low in all measured isotope ratios; they do not overlap with the rest of the Limni axis ores. Instead, they fall outside the compositional field of the known Mediterranean copper ores used in ancient times. The low measured isotope ratios would usually be held compatible with old deposits such as the Precambrian ores of the Arabian Peninsula or Oman (Artioli et al. 2020). One could suggest a similarity with the compositional field of some of the Wadi Arabah deposits (e.g. Ketelaer and Hauptmann 2016). However, an exact match with the Timna ores, for example (Segal et al. 2015) has not been found. It is unlikely that this unusual composition of the Pera Vasa slag is the result of mixing the signature of the ore with furnace materials: the lead isotopic composition of local clay and sediments appears to be in the same range as Cypriot copper ores, in many cases with higher ratios (e.g., Renson et al. 2013). If confirmed, this unusual signature of the Pera Vasa slag is in need of further investigation, not in the least because the lead isotope ratios for one of the metal samples coming from the excavation of Laona (LA2013.MAA17) is almost identical to one of the outlying slag samples from Pera Vasa. It should be noted that these samples do not differ in their chemical composition and that there are examples with both high and low manganese content with this unusual LIA fingerprint. The LIA of the Laona and Hadjiabdulla samples held more surprises. The second metallic sample from Laona (LA2013.MAA19) and the lump of high tin bronze from Hadjiabdulla (HA33-HA45_7_1) are both consistent with the ores from the area of Asgata-Kalavasos. This perhaps indicates that copper from that mining district, which is the third richest on the island and which probably belonged to the Iron Age polity of Amathus (Kassianidou 2013c), was exported to Paphos (Fig. 13).

Even more surprising is the fact that some of the slag samples from Laona seem to be consistent with the ores from the Solea mining district, which includes the mines of Apliki, Skouriotissa, Mavrovouni and Ambelikou. This phenomenon is not a unicum. Slag samples consistent with the Apliki ore deposit have been detected in the LBA sites of Kition (Hauptmann 2011), Enkomi (Gale and Stos Gale 2012) and Kalavasos-Agios Dhimitrios (Van Brempt 2016). It would appear that Paphos can now be added to the list.

4.4. *Anthracological study*

The anthracological study of organic material collected from the slag

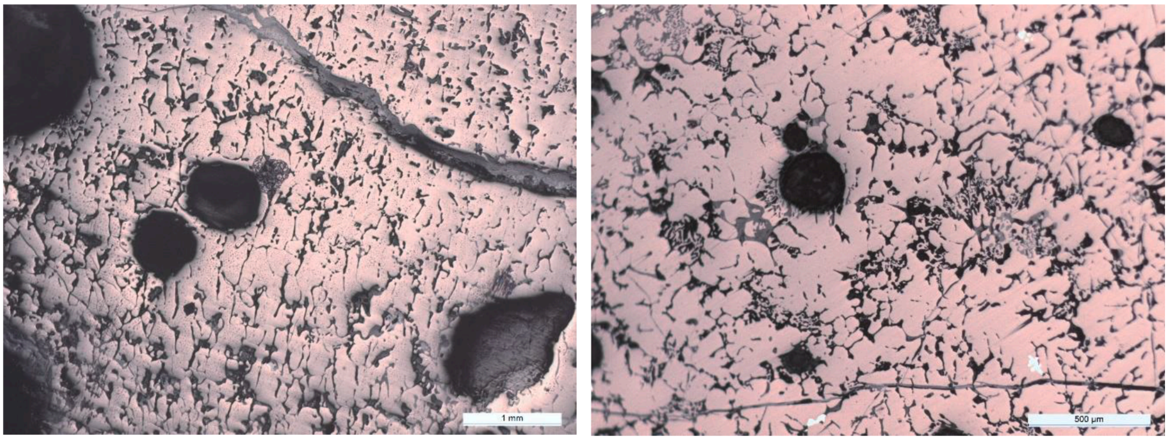

Fig. 11. Photomicrographs a: sample HA-21a (mag. 4x left) and b: HA-27 (mag. 10x right) showing hammer scale texture with small loss of ferrite (white globules).

Table 3
The results of the Lead Isotope Analysis.

Sample Number	Inventory Num.		^{206}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{208}Pb/^{204}Pb	^{207}Pb/^{206}Pb	^{208}Pb/^{206}Pb	^{208}Pb/^{207}Pb	σ^{206}Pb/^{204}Pb	σ^{207}Pb/^{204}Pb	σ^{208}Pb/^{204}Pb	σ^{207}Pb/^{206}Pb	σ^{208}Pb/^{206}Pb	σ^{208}Pb/^{207}Pb
1	LA2012.MAA6	Slag	17.738	15.550	37.568	0.877	2.118	2.416	0.003	0.003	0.006	0.000	0.000	0.000
4	LA2013.MAA6	Slag	18.483	15.600	38.424	0.844	2.079	2.463	0.003	0.003	0.008	0.000	0.000	0.000
9	LA2013.MAA11	Slag	18.651	15.602	38.550	0.837	2.067	2.471	0.003	0.003	0.006	0.000	0.000	0.000
55	LA2013.MAA17	Metal (Cu)	18.090	15.592	37.988	0.862	2.100	2.436	0.003	0.002	0.005	0.000	0.000	0.000
56	LA2013.MAA19	Metal (Cu-Sn)	18.966	15.687	39.007	0.827	2.057	2.487	0.003	0.002	0.006	0.000	0.000	0.000
15	LA2013B.MAA6	Slag	18.546	15.577	38.420	0.840	2.072	2.466	0.004	0.003	0.009	0.000	0.000	0.000
22	HA33_5_2	Slag	18.488	15.641	38.512	0.846	2.083	2.462	0.004	0.004	0.008	0.000	0.000	0.000
25	HA33_5_4	Slag	19.017	15.642	39.051	0.823	2.054	2.497	0.008	0.006	0.016	0.000	0.000	0.000
27	HA33_5_5	Slag	18.723	15.660	38.743	0.836	2.069	2.474	0.002	0.002	0.005	0.000	0.000	0.000
28	HA33_5_5	Slag	18.795	15.668	38.798	0.834	2.064	2.476	0.003	0.002	0.006	0.000	0.000	0.000
58	HA33-HA45_7_1	Metal (Cu-Sn)	18.841	15.656	38.727	0.831	2.055	2.474	0.002	0.002	0.005	0.000	0.000	0.000
30	PV Point 1 31a	Slag	18.614	15.671	38.713	0.842	2.080	2.470	0.005	0.004	0.010	0.000	0.000	0.000
33	PV Point 2– 32a	Slag	18.713	15.660	38.773	0.837	2.072	2.476	0.007	0.006	0.014	0.000	0.000	0.000
36	PV Point 3– 33a	Slag	18.349	15.727	38.476	0.857	2.097	2.446	0.006	0.006	0.014	0.000	0.000	0.000
39	PV Point 4– 34a	Slag	18.126	15.603	38.053	0.861	2.099	2.439	0.003	0.003	0.007	0.000	0.000	0.000
42	PV – 2022.1	Slag	16.928	15.480	36.675	0.914	2.167	2.369	0.004	0.003	0.009	0.000	0.000	0.000
43	PV 2022.2	Slag	18.303	15.638	38.426	0.854	2.099	2.457	0.010	0.009	0.025	0.000	0.000	0.000
45	PV 2022.4	Slag	18.794	15.838	39.097	0.843	2.080	2.469	0.030	0.026	0.065	0.000	0.000	0.000
46	PV 2022.5	Slag	17.581	15.608	38.518	0.842	2.078	2.468	0.017	0.015	0.040	0.000	0.000	0.000
47	AE – 35a (Point 1)	Slag	18.791	15.779	39.046	0.840	2.078	2.475	0.006	0.005	0.013	0.000	0.000	0.000
51	AE Point 2 – 36a	Slag	19.040	16.029	39.623	0.842	2.081	2.472	0.011	0.010	0.025	0.000	0.000	0.000
54	AE Point 3– 37	Slag	18.487	15.675	38.602	0.848	2.088	2.463	0.002	0.002	0.005	0.000	0.000	0.000

heaps of Pera Vasa and Agios Georgios Emnon revealed that *Pinus brutia* is the taxon present in all samples from both slag heaps. *Olea europaea* was also identified in one sample per site. The results are not unexpected. The sites are in, or close to, the Paphos pine forest and a similar vegetation would have occurred in the past. Similar results regarding the exploitation of pine woodland have been reached through wood charcoal analysis at other ancient smelting workshops and slag heaps in Cyprus (Fasnacht et al., 1991; Ntinou, 2013; Socratous et al. 2015). Pine wood was the chosen fuel for the smelting furnaces because the pine forest was readily available for the needs of the metallurgical activities. The understory of dense pine forests is rather poor in other ligneous plants but pine wood in different states (healthy standing trees, standing dead trees, fallen trees, dead wood in the form of dead or dying branches in the canopy, fallen lateral branches) is available in considerable quantities.

5. Conclusions

The chemical, microscopic and isotopic analysis of an archaeometallurgical assemblage consisting of slag and metallic samples from two Cypro-Classical monuments of the urban landscape of Paphos and from two slag heaps in the metalliferous region of Paphos has enabled a pioneering diachronic study of copper production in the area. Like in all the mining regions of Cyprus, the extent of the working in Late Antiquity hinders the visibility of earlier exploitation. Nevertheless, the LIA analysis suggests that these deposits were exploited in earlier periods; it also suggests that metal from the mines of Kalavasos and Asgata was imported to Paphos. The chronological horizon of the transportation is vague but the possibility that copper metal was exchanged between mining regions opens a new and challenging chapter in the industrial economies of the Cypriot polities; it also renders the identification of the copper fingerprint of Paphos even more urgent. In view of this, the

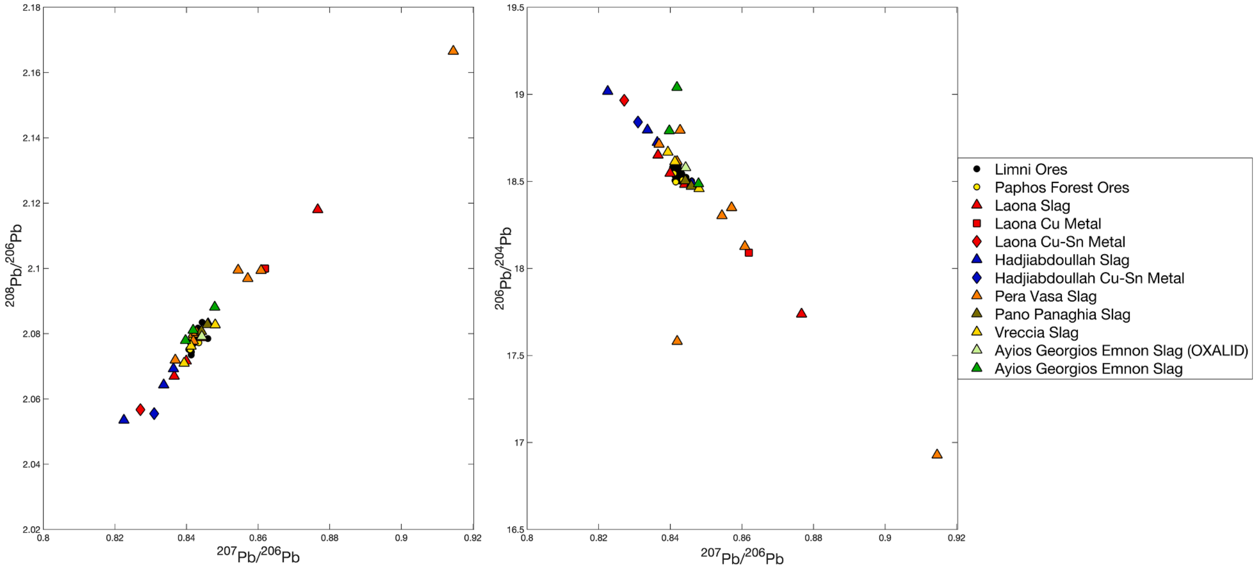

Fig. 12. Lead isotope analysis results plotted against data for ore and slag from Paphos area and Limni axis (from OXALID database of Cypriot ore and slag).

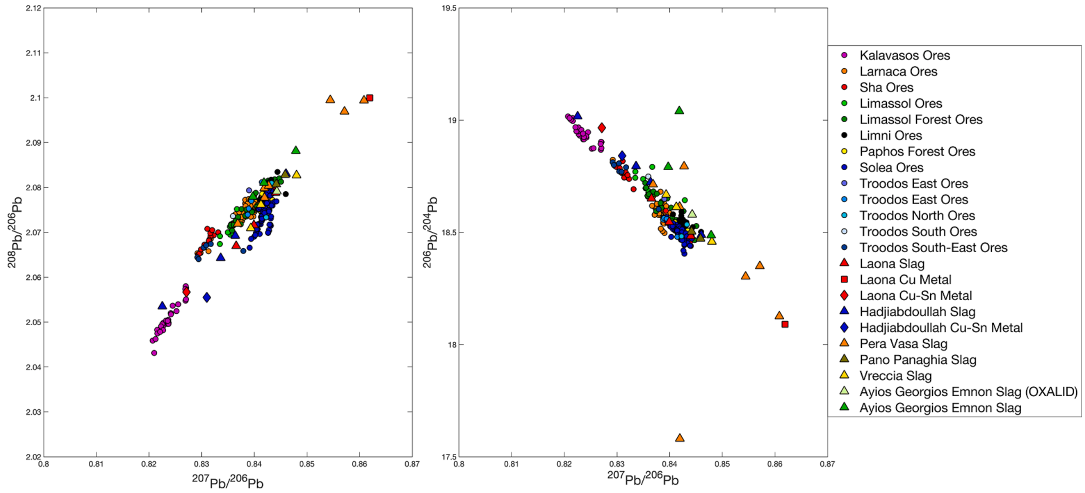

Fig. 13. Lead isotope analysis results (excluding the two outlier samples from Laona) plotted against all data for Cypriot ore and slag (from OXALID database of Cypriot ores and slag).

unusual slag and metal compositions, identified through LIA, which cannot be related to any known ore deposit, require further investigation. Technological changes and innovations, such as the introduction of new types of fluxes and the optimization of the smelting technology, were also identified, and in the workshop complex of the Paphian citadel the waste products revealed the installation of an iron smithy.

This study is a first but concise step towards the formal incorporation of the Paphos copper deposits in the on-going archaeo-metallurgical projects studying the copper of Cyprus. The chronological depth and the extent of the exploitation, as well as the fingerprint of the Paphos copper ores, are no longer beyond our grasp.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data is presented in the manuscript

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